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Burner Criteria for NOx Control. Volume I. Influence of Burner Variables on NOx in Pulverized Coal Flames

International Flame Research Foundation, Ijmuiden (Netherlands)

Prepared for

Industrial Environmental Research Lab, Research Triangle Park, N C

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BURNER CRITERIA FOR NO_x CONTROL
Volume I - Influence of Burner Variables on NO_x in Pulverized Coal Flames

UNITED STATES ENVIRONMENTAL PROTECTION AGENCY

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16. ABSTRACT The report gives results of the first phase of an investigation to specify burner design criteria to control NOx in natural gas and pulverized coal flames. The two parameters found to have major influence on NO formation were the method of fuel injection and the degree of swirl. NO formation can be controlled by optimizing burner design parameters because its rate of formation depends on the detailed mixing history of the fuel, combustion air, and recirculating combustion products. The same parameters also dictate such flame characteristics as stability, length, and luminosity. An explanation of the influence of burner parameters on pulverized coal flames is based on two assumptions: the most significant factor of the total emission is fuel NO, and the emission variation depends on the fate of the volatile nitrogen compounds. Fuel NO formation can be reduced by ensuring that the volatile nitrogen compounds react under oxygen deficient conditions. Maximum emissions occur with radial fuel injects because the coal is rapidly mixed with the total air supply and hot recirculating products. These conditions ensure early stable ignition. However, fuel/air mixing promotes NO formation. Conversely, NO formation can be restricted by maintaining the fuel in a coherent axial jet and discouraging primary/secondary mixing by surrounding the fuel jet with an ignition front. The coal must also be delivered with the minimum amount of primary air.

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VOLUME I. INFLUENCE OF BURNER VARIABLES ON NO_x IN PULVERIZED COAL FLAMES

by

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FOREWORD

This report presents the experimental results and initial evaluations of an investigation to develop burner design criteria to control the formation of nitrogen oxides in large scale turbulent diffusion flames. The investigation was carried out at the IJmuiden Research Station of the International Flame Research Foundation.

We wish to express our gratitude to former colleagues at the Research Station who did much of the groundwork for this investigation, particularly Dr. W. LEUCKEL, Dr. N. FRICKER and Dr. K. HEIN. We would also like to express our thanks to Professor J. BEER and members of the Programme Executive for their helpful advice and to the staff members of the Research Station for help in the preparation and execution of the experimental programme. We are also indebted to the many Foundation members who gave freely of their own experience.

Mr. G. B. MARTIN is the E.P.A. technical project officer whose invaluable contribution is gratefully acknowledged. Finally, we wish to thank Dr. E. BERKAU and Mr. D. W. PERSHING for the stimulating discussions.

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1. SUMMARY

Modern society's increasing demand for energy has created two problems of general importance for combustion engineers: the depletion of the world's reserves of fossil fuel, and the pollution of the atmosphere by combustion-generated pollutants. These problems are interlinked and should not be considered separately. This report includes the results obtained during the first phase of an investigation to specify burner design criteria to control NO_x in natural gas and pulverized coal flames. Parametric investigations have been carried out to determine the influence of burner parameters on NO formation in natural gas and pulverized coal flames.

The influence of the following burner parameters has been investigated:

- the method of fuel injection;
- the throat velocity;
- the geometry of the burner exit;
- the position of the fuel injector;
- the type of burner exit;
- the proportion of primary air; and
- the swirl intensity of the combustion air.

The two parameters found to have a major influence on NO formation were the method of fuel injection and the degree of swirl. Nitric oxide formation can be controlled by the optimization of burner design parameters because its rate of formation is determined by the detailed mixing history of the fuel, the combustion air and recirculating combustion products. The same parameters also dictate flame characteristics.

The flames investigated can be broadly classified into three groups:

1) Lifted flames - the ignition front is stable in space with time but it is established downstream from the fuel injector;

2) Simple jet flames - flames produced with axial fuel injection and without a swirl-induced internal recirculation zone; and

3) High intensity swirling flames - in Type I flames the fuel jet penetrates the internal recirculation vortex. In Type II flames the fuel jet is diverted around the recirculation vortex which forms on the flame axis. (See Section 3.2.1 for explanation of flame types.)

Figure 1.1 shows a composite diagram of the emission characteristics of the various types of natural gas flames. It can be seen that the emission characteristics of the various flame types overlap, this has important practical implications since it is possible to optimize the burner design to control pollutant formation and yet provide flame characteristics which satisfy process requirements. This was not observed with pulverized coal flames; maximum emissions were always obtained with high intensity Type II flames. However, minimum emissions were provided by axial jet flames and the emission level for these flames was comparable to emissions from natural gas flames with similar input conditions.

Figure 1.2 gives an indication of the range of emission characteristics which were observed with coal flames when two parameters, fuel injector and swirl level were varied. The characteristics shown in Figure 1.2 have several interesting features:

1) The step change in emission level with increasing swirl observed with injector A. This was due to the establishment of an ignition zone at the fuel injector.

2) The high emission level flames produced with the radial fuel injector (F) and the almost lack of dependence on swirl level.

3) The low emissions measured with high velocity, single hole axial fuel injection (H).

In order to explain these observations two basic assumptions have been made. These are that the most significant factor of the total emission is fuel NO and that the variation in the emission depends upon the fate of the volatile nitrogen compounds. Fuel NO formation can be reduced by ensuring that the volatile nitrogen compounds react under oxygen-deficient conditions. Maximum emissions occur with radial fuel injection because the coal is rapidly mixed with the total air supply and hot recirculating products. These conditions ensure early stable ignition which is an important practical requirement. However, the fuel/air mixing promotes NO formation.

Conversely, NO formation can be restricted by maintaining the fuel in a coherent axial jet and discouraging primary/secondary mixing by surrounding the

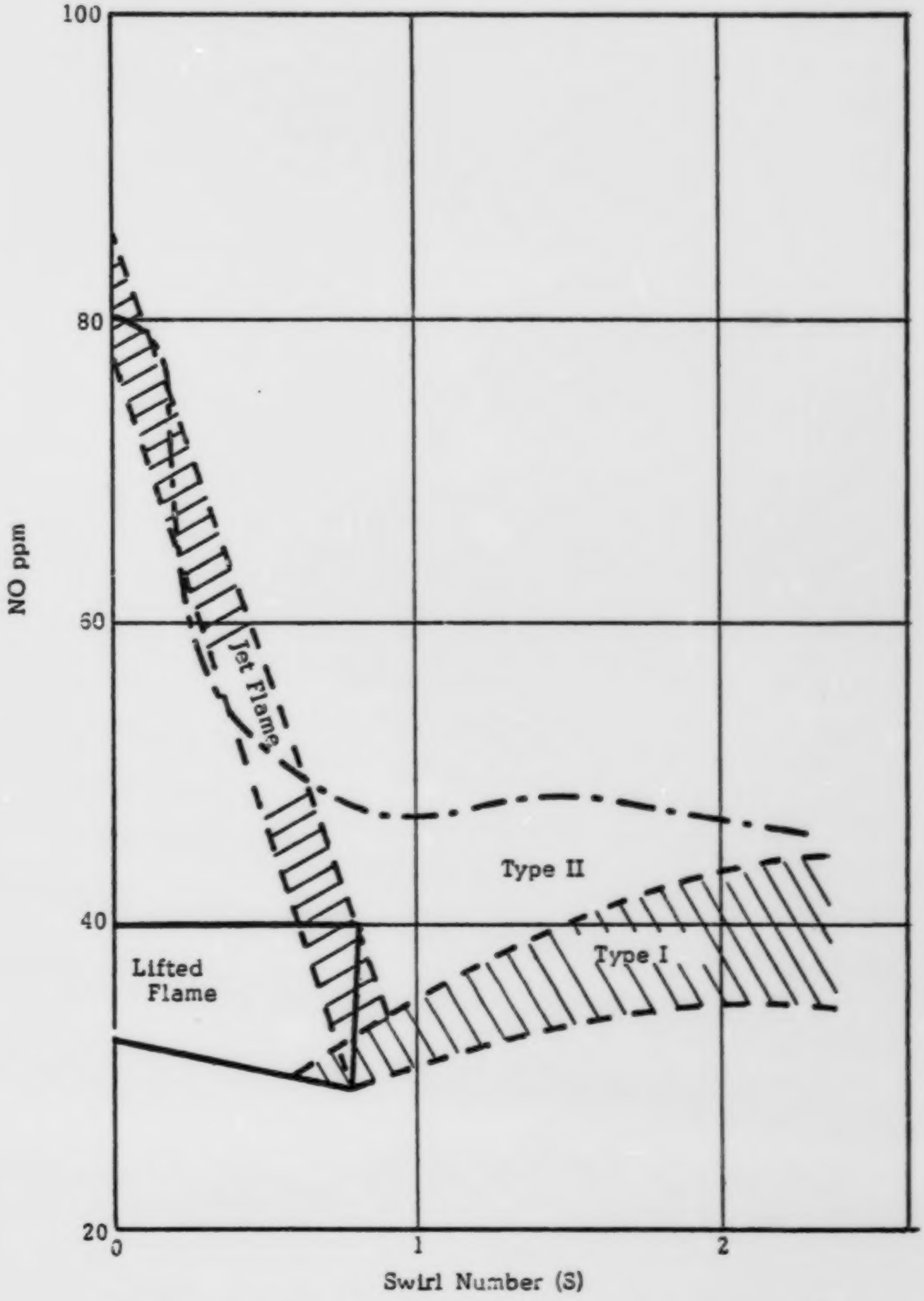

Figure 1.1 NO Emission Characteristics Of The Various Flame Types

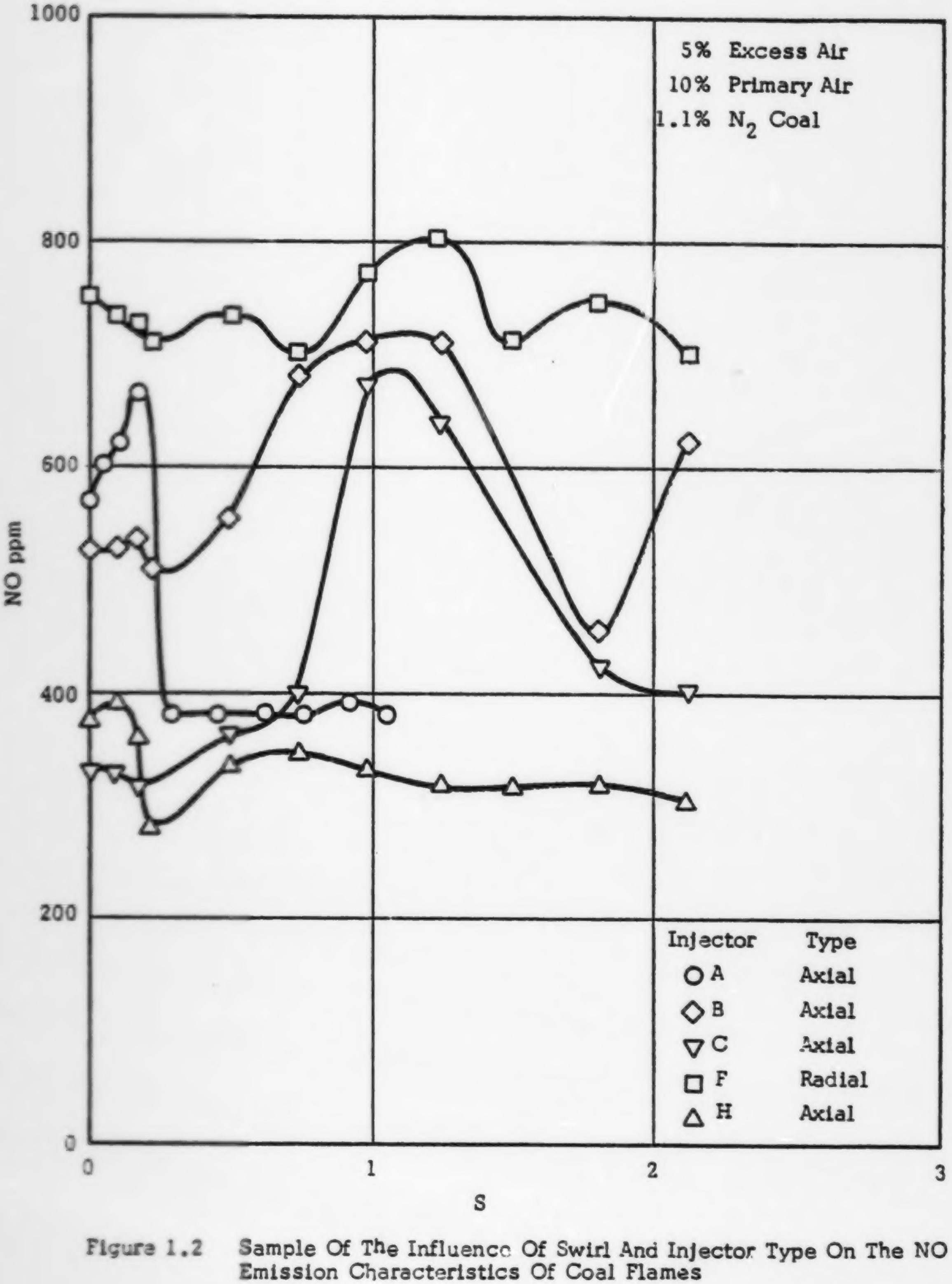

Figure 1.2 Sample Of The Influence Of Swirl And Injector Type On The NO Emission Characteristics Of Coal Flames

fuel jet with an ignition front. The fuel must also be delivered with the minimum amount of primary air.

Unfortunately these conditions produce flame characteristics which are incompatible with existing boiler designs. Future work will concentrate upon providing flame characteristics which satisfy process requirements and yet have minimum emissions.

2. PURPOSE AND SCOPE OF THE RESEARCH PROGRAM

Nitrogen oxides are produced during the combustion of all fossil fuels using air as the oxidant and the recognition (2.1) of their significance as atmospheric pollutants has resulted in public pressure to control their emission into the atmosphere. It has been estimated (2.2) that stationary combustion sources accounted for 50% of the 23 million tons of NO_x emitted in the United States in 1972. The objective of this work is to provide criteria to enable burners to be designed which will allow fossil fuels to be utilized efficiently with the minimum pollutant emission.

Various control techniques are available to reduce NO_x emissions from large steam raising plants (2.3, and 2.4). These techniques include:

- operating modifications e.g., reduced load, excess air or preheat;
- combustion modifications e.g., flue gas recirculation or staged combustion;
- burner redesign or modification.

All these techniques will necessitate variations in the accepted plant operating conditions which may reduce the boiler efficiency. In the long term burner design may well prove to be the most efficient method of controlling pollutant emission from all forms of fossil fuel fired furnaces and boilers.

This report covers Phase I of a research program whose overall objective is the specification of burner design criteria for minimum pollutant emissions from p.f. and heavy oil fired furnaces. The program was divided originally into three phases and the scope of the work for each phase is summarized below.

Phase I

1. Construct and evaluate a system capable of withdrawing and analyzing gas smaples from natural gas and p.f. flames for nitrogen oxides.
2. Determine the effect of burner parameters on emissions from gas and p.f. flames.

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3. Make detailed temperature, concentration and velocity maps of several gas and coal flames.
4. Inititate a program which will allow the amount of nitrogen oxides emitted by natural gas and p.f. flames to be predicted.

Phase II

1. Investigate the effect of burner parameters on nitrogen oxide emissions from fuel oil flame.
2. Investigate the effect of the following variables on nitric oxide emission from fossil fuel flames:
    - air preheat level;
    - firing rate;
    - nitrogen content of the fuel.
3. Continue the work begun in Phase I to develop a predictive model.

Phase III

1. Either design and construct a furnace to investigate the effect of rate of heat loss on nitrogen oxide formation in flames or continue promising lines of research indicated by the earlier work.
2. Continue the development of the predictive model.

This report presents the results of Phase I and attempts to explain the apparently contradictory information obtained on the effect of burner parameters on nitric oxide formation.

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2.2 "Air Quality and Stationary Source Emission Control", a Report prepared for the Committee on Public Works, U.S. Senate, March 1975.

2.3 Bartok, W., Crawford, A.R., Cunningham, A.R., Hall, H.J., Manny, E.H. and Skopp, A., "Systems Study of Nitrogen Oxide Control Methods for Stationary Sources". Final report, Contract No. PH-22-68-55, PB 192789, 1969.

2.4 Breen, B.P., "Emissions from Continuous Combustion Systems". Ed. by W. Cornelius and W. G. Agnew, p. 325, Plenum Publishing Corporation, New York, 1972.

3. BACKROUND

3.1 Nitric Oxide Formation In Fossil Fuel Flames

The parametric studies described below demonstrate the strong influence of burner variables on nitric oxide formation in natural gas and pulverized coal flames. The nitric oxide emitted from fossil fuel flames originates from two sources:

- the oxidation of molecular nitrogen, termed thermal NO;
- the conversion of nitrogen compounds contained in the fuel, termed fuel NO;

In order to understand the effect of burner variables on NO emissions it is necessary to review the factors which control the formation of both thermal and fuel NO in flames.

3.1.1 The Formation of Thermal NO

It is generally agreed that the principal reactions involved in the oxidation of molecular nitrogen flames are (3.1 - 3.5):

$$N_2 + O \longrightarrow NO + N \qquad (1)$$

$$N + O_2 \longrightarrow NO + O \qquad (2)$$

$$N + OH \longrightarrow NO + H \qquad (3)$$

and that NO formation rates in post flame gases can be successfully modelled with these reactions assuming equilibration of the oxygen atoms. Lavoie et al (3.6) have suggested that in addition reactions (4) to (6) should also be considered

$$OH + N_2 \longrightarrow N_2O + H \qquad (4)$$

$$O + N_2O \longrightarrow NO + NO \qquad (5)$$

$$O + N_2O \longrightarrow N_2 + O_2 \qquad (6)$$

However, Bowman (3.2) and Westenberg (3.3) consider that in practical cases reactions involving N_2O do not play a significant role in the formation mechanism.

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It has been reported (3.2, 3.5 and 3.7) that nitric oxide formation rates measured in the vicinity of flame fronts exceed the rates predicted by reaction (1) - (3). Fenimore (3.5) refers to this rapidly formed nitric oxide as "prompt" NO and considers that its formation is due to the destruction of the $N \equiv N$ bond by reactions other than those shown above. Since the formation of "prompt" NO was restricted to hydrocarbon mixtures, Fenimore suggested that the formation of HCN or CN by reactions such as

$$CH_2 + N_2 \longrightarrow CHN + \ldots. \tag{7}$$

$$C_2 + N_2 \longrightarrow CN + CN \tag{8}$$

followed by oxidation of the CN could easily lead to nitric oxide formation.

Several workers (3.2 and 3.8) consider that the increased rate of formation of nitric oxide in the vicinity of flame fronts is attributable to non equilibrium radical concentrations. In shock tube studies Bowman and Seery (3.4) were able to predict measured rates of nitric oxide formation with reactions (1) - (3) provided the O and OH concentrations were correctly evaluated De Soete (3.8) also considers that the concentration of free radicals will exceed equilibrium levels near flame fronts. De Soete found that the overall activation energy for the formation of NO in combustion products was dependent on residence time and varied from values as low as 87 kcal/mole for short times and increased to 135 kcal/mole as the residence time increased.

The controlling influences of time and temperature on the formation of nitric oxide in combustion products are aptly demonstrated in the work of Breen et al (3.10). The rate of thermal NO formation is a maximum for adiabatic, stoichiometric combustion and rapidly decreases as the fuel air mixture ratio varies from stoichiometric. Consequently, all the techniques which have been used to control thermal NO formation (3.11 and 3.12) attempt to reduce peak flame temperatures and residence times in high temperature zone.

3.1.2 The Formation of Fuel NO

It is universally accepted that the oxidation of nitrogen compounds in fossil fuels contributes significantly to the total nitric oxide emission from flames. However, the conversion of fuel nitrogen compounds to nitric oxide in flames has not been studied as extensively as the formation of thermal NO. It is known that residual fuel oils and coal contain bound nitrogen. The nitrogen content of bitumenous coal varies in the range of 1.0% to 2.1%. Typical nitrogen containing compounds which have been isolated from coal by solvent extraction and temperature vacuum distillation include 2 and 4 methylpyridine, trimethylpyridine, quinoline, 2 methylquinoline, aniline and toluidine (3.13).

At the time of writing almost all the available information concerning fuel nitrogen conversion has been obtained with laboratory scale experiments using additives and doped fuels. Only limited information is available from large scale practical units. Bartok et al (3.14) estimate that the presence of 1% N in the fuel oil of a 175 M.W. front walled fired utility boiler increase NO emission from a baseline 320 to 580 ppm. In a 320 M.W. tangential fired unit 1% nitrogen concentration resulted in an increase in the flue gas nitric oxide content of 140 ppm from a baseline of 150 ppm. Emission of 400 ppm NO have been reported of a coal fired fluidized bed combustor using argon oxygen mixtures instead of air for combustion (3.15). Essentially all the nitric oxide emitted was derived from fuel nitrogen, bed temperatures were of the order of 800°C and it was found that emissions increased as the oxygen content of the "argon air" was increased.

Shaw and Thomas (3.16) have reported the conversion of gas phase nitrogen compounds including pyridine, to NO in low temperature premixed rich carbon monoxide flames. Nitric oxide was produced whether the oxygen and fuel were mixed with either nitrogen or argon.

Bartok, Engelman and del Valle have studied the effect of the addition of various nitrogen compounds to a Longwell combustor (3.17). This combustor allows examination of kinetically-limited combustion phenomena where transport effects are minimized. The compounds investigated were NO, NO_2, NH_3, CH_3-NH_2 and C_2N_2 and results can be summarized as:

- conversion to or retention of NO decreases as the combustion mixture becomes fuel-rich;
- complete conversion of NO and NO_2 was noted with fuel-lean mixtures;
- the percent conversion of NH_3 to NO decreased as the NH_3 concentration increased; and
- complete conversion of cyanogen was never achieved.

One of the most significant results reported by Bartok et al was that nitric oxide could be reduced to molecular nitrogen (indicated by absence of NO in combustion products) in fuel-rich combustion conditions. Also a recent patent of the John Zinc Company (3.18) is concerned with the homogeneous conversion of NO to N_2 by combustion in a fuel-rich environment.

Martin and Berkau (3.19) and Turner et al (3.20) have investigated the conversion of fuel nitrogen compounds in combustion systems using doped distillate fuels. However, both workers used different techniques to separate the thermal NO_x from the fuel nitrogen NO_x. Martin assumes that for any given condition the thermal NO_x is the emission from the undoped fuel. Turner et al postulated that flue gas recirculation only reduced thermal NO and that the elimination of all thermal NO formation could be achieved by 30 percent recirculation. This postulate was based on the fact that 30 percent recirculation reduced the emission of NO from a No. 2 oil (0.03 percent N_2) to 40 ppm with both high and low atomizing air flow rates. The emission of 40 ppm was exactly that amount which would be expected from total conversion of the fuel nitrogen. With doped fuels 30 percent recirculation also reduced emissions by the same amount.

The assumptions used by both Martin and Turner to allow them to determine the amount of fuel NO produced with doped fuels are open to criticism. The nitrogen additive may influence thermal NO formation by altering the surface tension or the viscosity of the fuel. Flue gas recirculation of 30% does not entirely eliminate NO formation from gas fired systems and the basis for Turner's assumptions may be due to some peculiarity of his particular experimental system. A major criticism of experiments with doped fuels is associated with the type of nitrogen bonds within the fuel. However these experiments have given important indications of the behaviour of nitrogen compounds in real fuels.

Both Martin and Turner found that the fraction of fuel nitrogen converted to nitric oxide increases with increasing excess air and decreases with fuel nitrogen concentration. Martin found that quinoline (C_9H_7N) gave lower conversions than piperdine ($C_5H_{11}N$) and pyridine (C_5H_5N). The latter two gave identical results. Turner found little influence of the boiling point of the nitrogen additive with twenty different compounds in a No. 2 oil. Only when the b.p. was less than 100°C did the conversion to fuel NO decrease considerably.

The work of Martin and Turner produced similar general conclusions but there are detailed differences in their results. Turner found greater conversions than Martin suggesting that the formation of fuel NO is a strong function of the burner/ combustion chamber system. This conclusion is supported by Shell data reported by Sternling and Wendt (3.21). Emissions of 543 ppm NO_x and 300 ppm NO_x were measured from two different burners firing the same oil into two similar heaters. These workers also report that inefficient atomization systems tend to give lower nitrogen fuel at the same excess air level.

Ammonia is the most simple fuel molecule containing chemically bound nitrogen and it could be formed by pyrolysis of a more complicated molecule. Approximately 16% of the nitrogen in coal is converted to ammonia during conventional carbonization processes. Detailed species concentration measurements in low pressure ammonia flames (3.22) have shown that the nitric oxide concentration increases rapidly in the flame zone to values significantly greater than the equilibrium values. The NO concentration then decreases in the post flame zone.

The rate of disappearance of NO is relatively slow in lean flames but can be quite rapid in rich flames. Fenimore and Jones (3.22) proposed that NO was reduced by NH_2:

$$NO + NH_2 \rightarrow N_2 + \ldots \tag{9}$$

whereas Drummond and Hiscock (see Ref. 3.21) account for the reduction by the reaction:

$$NO + NH \rightarrow N_2 + OH \tag{10}$$

Bowman (3.2) has recently proposed a partial equilibrium mechanism to account for nitric oxide formation in shock-induced combustion of lean hydrogen, oxygen and ammonia mixtures. Bowman considers that nitric oxide is produced by the rapid reaction of O or OH with the fuel nitrogen intermediate. Subsequent reaction of NO will then occur through relatively slow reactions such as:

$$O + NO \rightarrow N + O_2 \tag{11}$$

$$NO + NO \rightarrow N_2O + O \tag{12}$$

$$NO + RN \rightarrow \text{....eventually } N_2 \tag{13}$$

Consequently the non-equilibrium NO concentrations which are observed in the combustion zone are the consequence of non-equilibrium radical concentrations in that zone.

Sternling and Wendt (3.21) have attempted to develop a kinetic model which will take account of the experimental results obtained with doped fuels. They consider that the model should explain the following:

- the fractional conversion of fuel nitrogen to NO decreases as the quantity of fuel nitrogen increases;
- the conversion of fuel nitrogen to NO increases with increasing excess air;
- under rich flame condition NO can be reduced to N_2.

Sternling and Wendt consider that the Zeldovich mechanism alone is not adequate to explain the reduction of NO under fuel rich conditions. The nitrogen bond is formed from the reaction

$$N + NO \longrightarrow N_2 + O \qquad (14)$$

which is faster under fuel rich conditions than

$$N + O_2 \longrightarrow NO + O \qquad (15)$$

The nitrogen atoms are built up by reactions involving HNO, NH etc.

Fenimore (3.23) has recently reported experiments in which he added nitrogen compounds to premixed ethylene flames. Fenimore considers that all fuel nitrogen compounds are converted to an intermediate compound I which subsequently reacts to produce either NO or N_2:

$$I + R \longrightarrow NO + \ldots. \qquad (16)$$

$$I + NO \longrightarrow N_2 + \ldots. \qquad (17)$$

Fenimore suggests that I could be either NH_2 or N.

Experiments both with doped fuels and simple nitrogen compounds suggest that the formation of fuel NO is strongly dependent on the combustion conditions under which the nitrogen compounds react. All the studies indicate that the formation of fuel NO can be reduced by restricting the available oxygen during the combustion of the fuel nitrogen compounds.

3.2 The Characteristics of Swirling Flames

Ignition stability is of paramount importance for all industrial burner systems. Two mechanisms of flame stabilization can be distinguished:

- stability is achieved because the tendency for the flame to propagate is in equilibrium with the flow of reactants, e.g., a simple premixed flame; and
- stability is achieved by the continuous ignition of a combustible mixture by mixing with hot gases which have been produced as a result of an earlier ignition, e.g., a bluff body stabilized flame.

All the flames discussed in this report were stabilized by the latter mechanism. A combustible mixture is mixed with hot recirculating gases, combustion is initiated and a flame propagates throughout the mixture. This flame may then act as an ignition source enabling the combustion of the fuel to take place and to maintain a stable ignition zone at the burner. Should the feedback of heat by the recirculating gases be insufficient to maintain a stable ignition source the flame will move downstream from the burner.

From the above description it can be seen that the presence of a recirculation zone is necessary to ensure adequate flame stability. The size of the recirculation zone and the temperature and velocity of the recirculating gases must be such that ignition is assured to maintain the recirculation zone in a condition necessary to assure subsequent ignition. Two types of recirculation zones are found in combustors:

- external recirculation zones; recirculation takes place between the boundary of the flame and the combustor walls. Although matter is entrained close to the burner, it originates further downstream; and
- internal recirculation; recirculation takes place within the flame boundaries and can be induced in the wake of a bluff body, or by imposing a strong axial gradient of static pressure at the center of the jet close to the burner. This static pressure gradient can be obtained by imparting a swirling tangential velocity component (swirl) to the jet fluid.

Swirl induced internal recirculation was utilized to ensure ignition stability for the majority of flames discussed in this report. The burner systems used during the investigation can be described as a double concentric jet burner with axial fuel injection and an annular variable swirling air flow.

The major effects of swirl are:

- reduced flame length;
- increased entrainment rates;
- improved stability.

The intraction of the axial fuel jet and the swirl induced torroidal vortex produce characteristic flame types. The swirl level necessary to produce a particular flame type depends very strongly upon the method of fuel injection. The condition necessary to produce these flame types and their properties will be described later.

The parameter most commonly used to characterize swirling flows is the swirl number S, defined by reference 3.24 as:

$$S = \frac{G_o}{G_x R} \qquad (18)$$

where:

G_o is the axial flux of angular momentum;
G_x is the axial flux of linear momentum;
R is the burner throat radius.

Leuckel (3.25) has compared, measured and calculated swirl parameters for the moving block swirl burner and these results are reproduced in Fig. 3.1. Figure 3.2 shows the relationship between block setting, R_s and swirl number for combinations of primary injector and throat diameters.

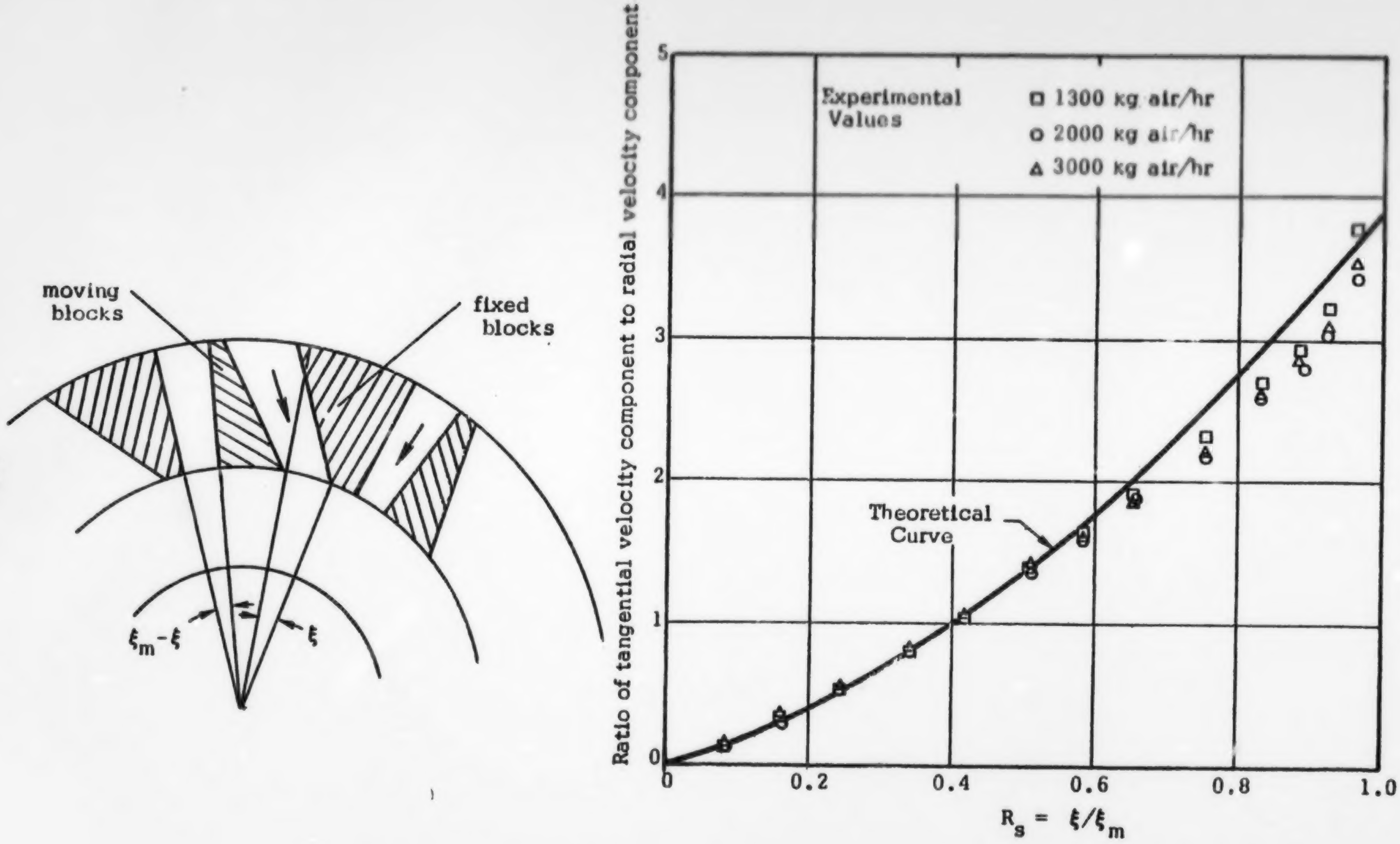


Figure 3.1 Movable-Block Type Swirl Generator

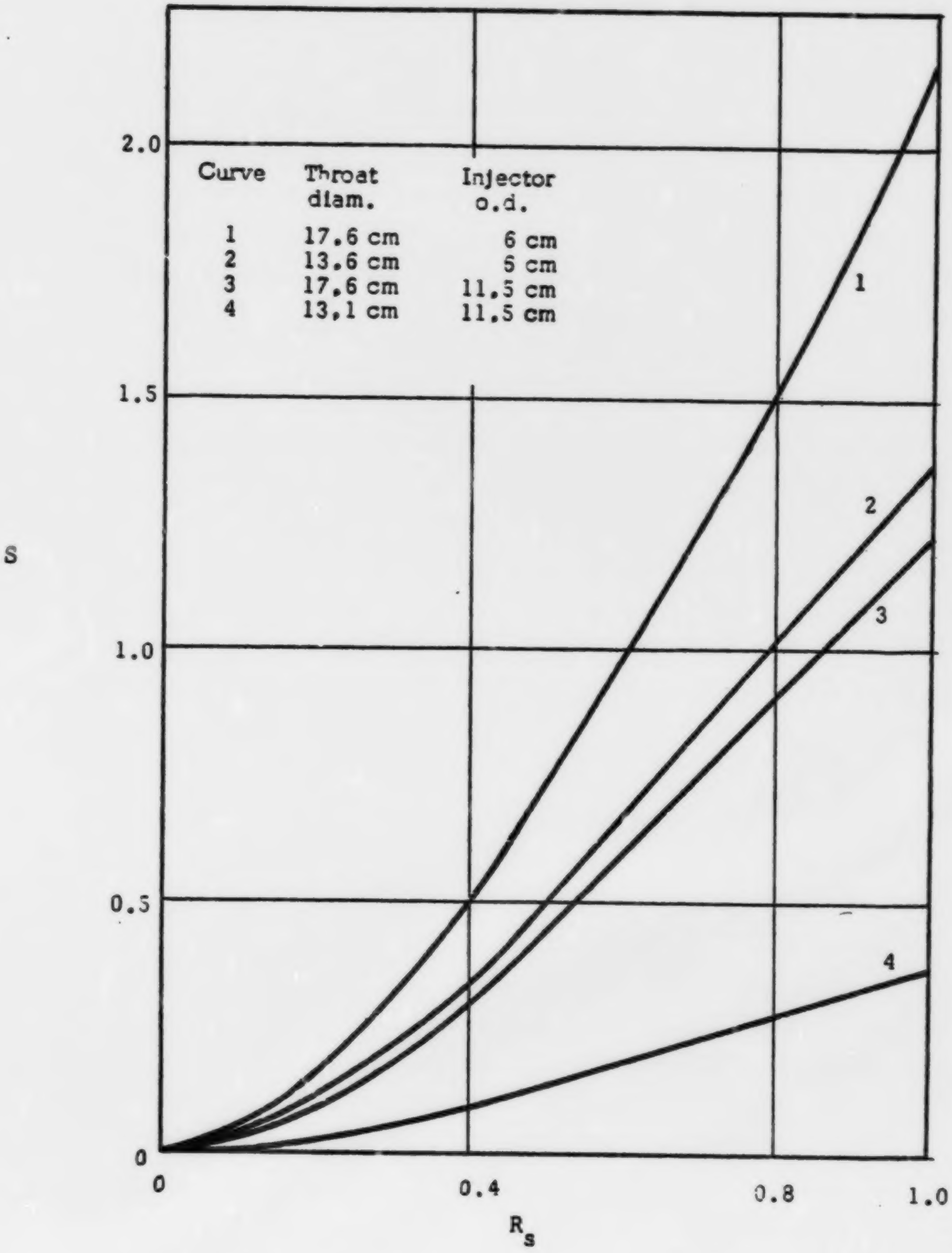


Figure 3.2 The Relationship Between Relative Swirl Index R_s And Swirl Number S

3.2.1 Classification of Swirling Flames

Swirl increases the mixing rate between the fuel and air close to the burner and at the boundaries of the recirculation zones (3.24 and 3.26). Consequently, flame length is reduced and combustion intensity (in terms of flame volume) is increased. The flame types described in this report can be conveniently classified in terms of combustion intensity. The flame sketches shown in Fig. 3.3 illustrate this simple classification scheme.

3.2.1.1 Low Intensity Jet Flames (Fig. 3.3a)

The swirl level is zero, or below that necessary to form an internal recirculation vortex, i.e. $S=0,6$. The flame could be either stabilized by a flame retention device (e.g. pilot flame) or by the recirculation zone formed at the interface between the primary and secondary streams. These flames are long and are characterised by low mean temperatures and slow increases in axial temperature. Increasing swirl from zero to $S = 0,6$ shortens the flame and increases the gradient of axial temperature rise.

3.2.1.2 High Intensity Type I Flames (Fig. 3.3b)

Increasing the swirl intensity beyond the critical forms an internal recirculation vortex close to the burner. If the momentum of the fuel jet is sufficient, then the internal recirculation zone is split by the fuel jet.

The resulting flame is divided into two sections:

- a short bulbous zone close to the burner;
- a longer tail section.

The two sections are connected by a short neck although under certain circumstance this connection may consist of only a few fillaments of flame which may be severed leaving only the bulbous front section. Alternately the two sections may merge and be almost indistinguishable. These flames are rarely encountered in practical systems.

3.2.1.3 High Intensity Type II Flames (Fig. 3.3c)

This type of flame is characterized by an internal recirculation zone on the flame axis and can be produced by:

- radial or wide angle divergent injection at low or zero swirl. At zero swirl the recirculation is due to the blockage caused by the fuel injector;
- divergent, annular or low momentum axial fuel injection at medium swirl levels;
- high momentum axial fuel injection at high swirl numbers.

Type II flames have a higher intensity than Type I flames and they can be produced at low swirl levels with the appropriate fuel injection system. Consequently, this is the type of flame is generally used in large steam raising boilers.

3.2.1.4 High Intensity Type III Flames (3.3d)

In addition to the two distinct flame forms referred to as Type I and Type II an intermediate form can occur. This flame has the bulbous base, characteristic of Type I flames but the tail is absent. This type of flame is associated with low annular air velocities and axial or annular fuel injectors. With high air velocities the change from Type I to Type III flames with increasing swirl is abrupt. The axial fuel jet enters the internal flow zone, but fails to achieve complete penetration and stagnates.

3.2.1.5 Lifted Flames (Fig. 3.3e)

Lifted flames are those where the ignition front is stable some distance downstream from the fuel injector. Increasing the swirl intensity normally produces a burner stabilized flame. The stability of lifted flames is enhanced by high external recirculation temperatures.

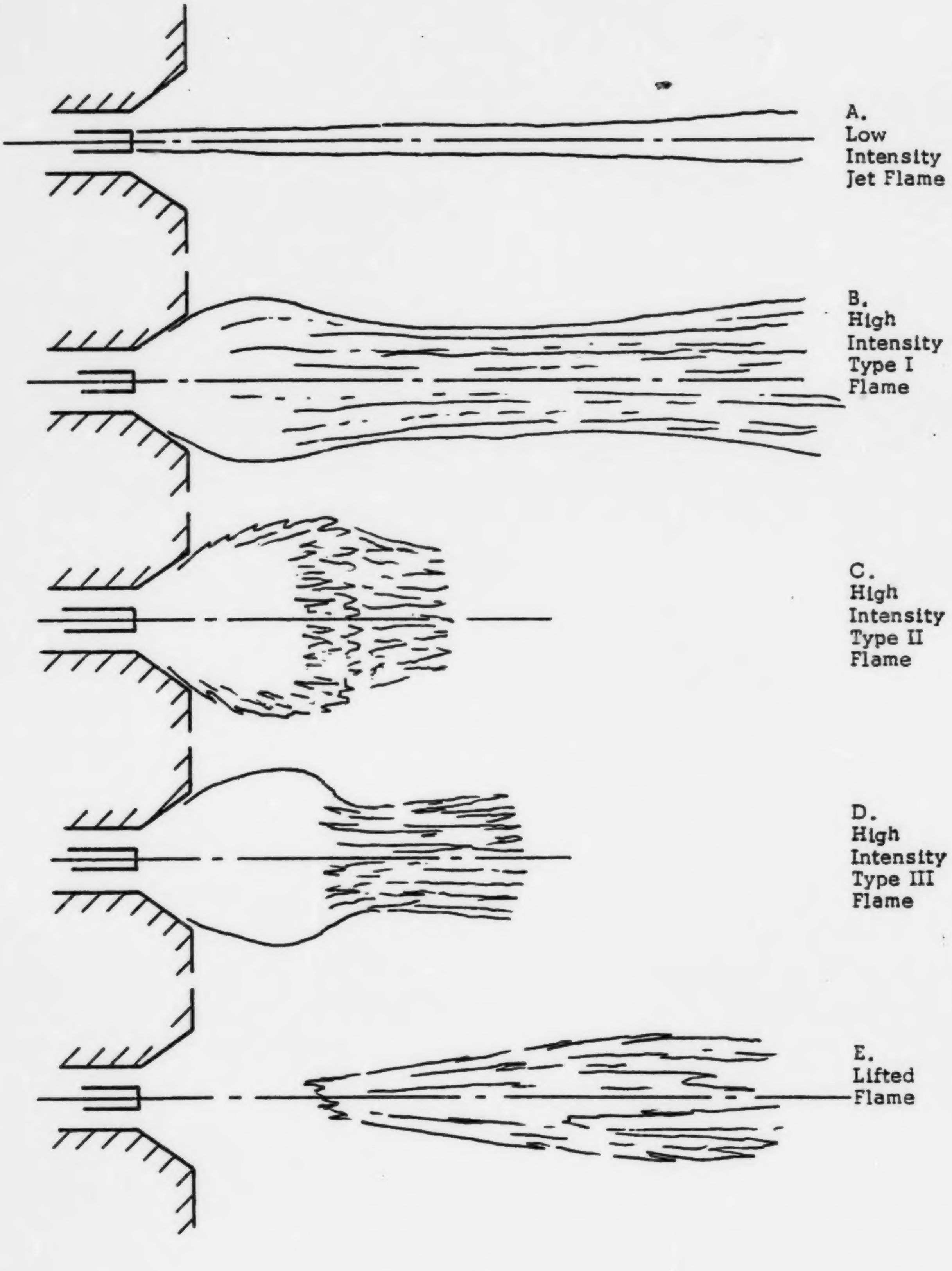


Figure 3.3 Simple Flame Classification Scheme

3.2.2 The Effect of Fuel Type

The four different flame types illustrated in Fig. 3.3 can be distinguished with either gaseous, liquid or solid fuels. This report is concerned with two fuels, natural gas and pulverized coal. In this section the effect of fuel type on the combustion characteristics of swirling flames will be discussed.

3.2.2.1 The Characteristics of Swirling Natural Gas Flames

The characteristics of swirling natural gas flames produced with a double concentric burner have been studied extensively by Leuckel and Fricker (3.27) and the following discussion relies heavily upon their work.

Conditions Necessary to Form Type I or Type II Flames

Several workers have established the importance of the parameter:

$$G = \frac{\text{primary jet velocity}}{\text{mean axial velocity of annular flow}}$$

in describing flow patterns in double concentric jets. The critical value of the parameter, G_c, at which the flame form changes from Type I to Type II is a function of several variables:

$$G_c = f\,(S, V_o\,(r), \alpha, L/D, \frac{x}{D}, N, f_g, f) \qquad (19)$$

where S = swirl number
V_o = distribution of tangential velocity
α = angle of burner divergent
L = length of burner divergent
D = diameter of throat
x = position of fuel injector within the divergent
N = factor representing injector type
f_g = fuel density
f = a factor to take account of the density distribution within the burner divergent and close to the exit

The last factor is an attempt to include a dependency on the "progress of combustion". The flow field in the region close to the burner will be strongly dependent on the static under-pressure developed by the swirling flow. The distribution of static pressure is given by

$$\frac{dp}{dr} = p\frac{V_o^{2}}{r} \qquad (20)$$

where V_o = tangential velocity at radius r

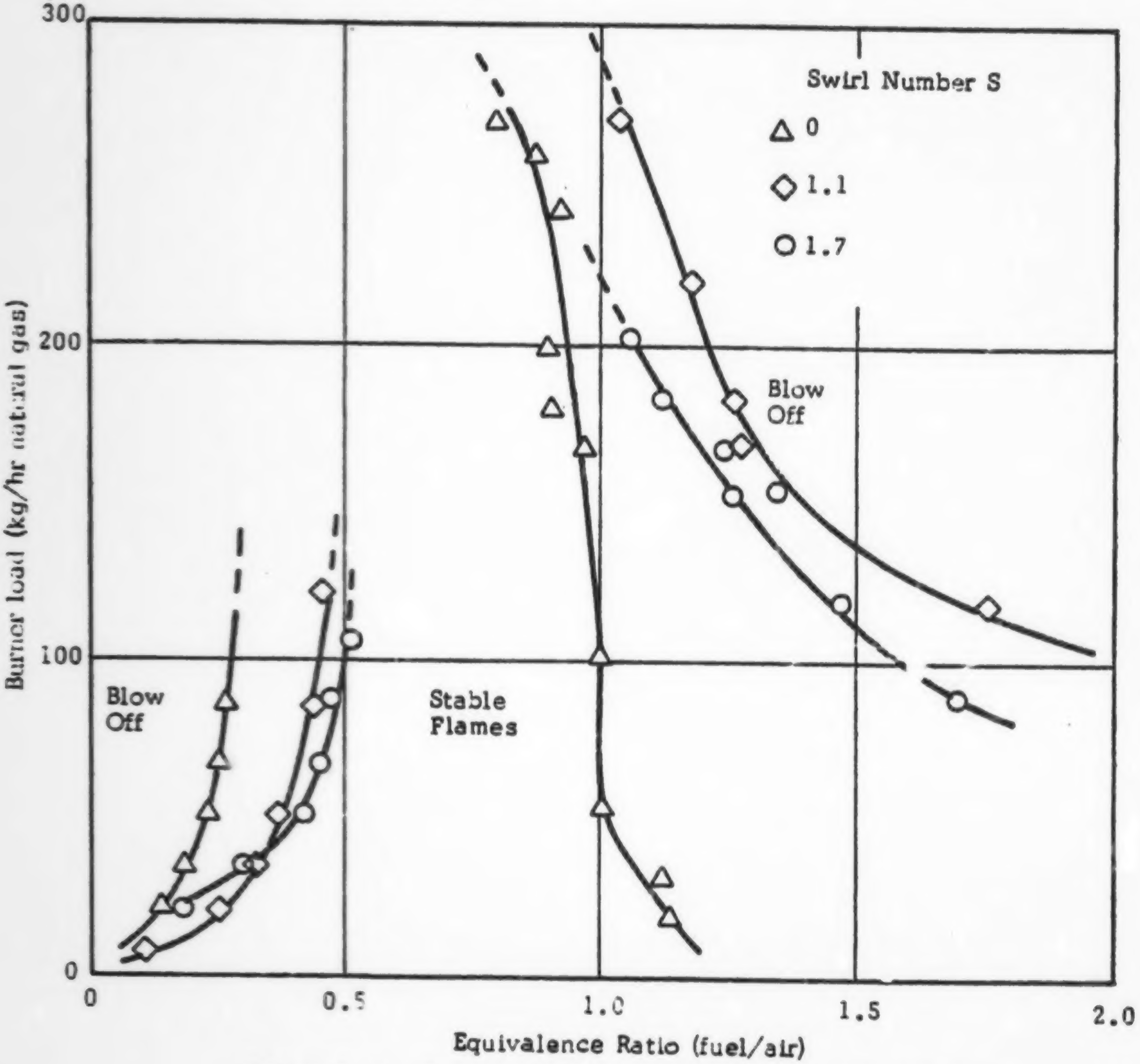
p = the density at radius r

Consequently, the static pressure field generated by a given distribution of tangential velocity will vary with the density and therefore the temperature of the swirling jet.

The relationship expressed in equation (19) contains the factor N to take an account of the method of fuel injection. With axial fuel injection low values of G promote the formation of Type II flames whereas high values of G allow the formation of Type I flames. When $G \simeq G_c$ the flame form fluctutates between the two stable forms. With annular fuel injection G_c is higher than for axial injectors, while with divergent gas injection it may not be possible to produce Type I flames. The effect of moving the fuel injector towards the exit of the burner divergent is to reduce the critical value of G_c. This can be explained by considering that the movement of the fuel injector towards the exit allows the gas jet to overcome part of the static pressure gradient without the associated loss in momentum of moving from the burner throat to the exit. Experiments have shown that divergent angles smaller than 15° encourage the formation of Type II flames (i.e., G_c is high). However, angles between 15° and 35° were not found to have an appreciable effect on flame type.

The Stability Characteristics of Swirling Natural Gas Flames

One of the benefits associated with the use of swirling flames is the greatly enhanced stability limits. Fig. 3.4 shows both fuel rich and fuel lean blow-off limit as a function of burner load at three swirl levels. It can be seen



Conditions

α = 25°
L/D = 1
Gas injector, annular (No. 21)
Gas velocity 35 m/s (166 kg/h)
Gas injected at burner throat

Figure 3.4 Region of Stable Combustion for Type II Flame (ref. 3.27)

that increased swirl widens the stability limits, in particular the rich limit. The improvement in flame stability is probably associated with two factors:

- improved fuel air mixing;
- an increase in the mass recirculated products to the flame base.

With premixed flames firing in the open stability limits can be reduced at high swirl levels. If the recirculation zone becomes too long, cold gases are returned to the base of the flames (3.28). Syred and Beer (3.26) were able to improve the stability limits of premixed flames by ensuring that short recirculation zones were formed.

The blow-off limits measured with the moving block swirl burner are dependent not only on swirl level, but also on the method of fuel injection and the burner geometry. Fricker (3.27) reports that:

- for flame stability an optimum divergent angle of 25° was found for both Type I and Type II flames. Angles less than 15° seriously reduced the burner load;
- for Type II flames there are little advantage in terms of flame stability in increasing the length of the burner divergent. However, when the fuel was injected from the throat the stability of Type I flames was improved by increasing the length of the divergent exit;
- with Type I flames blow-off could be attributed to excessive gas velocities. With Type II flame blow-off occurred if the annular air velocity was too high;
- for weak swirl the size of the interface between the fuel and air streams influenced the stability;
- divergent multihole or annular fuel injection improved the stability of Type II flames in comparison to those flames produced with low velocity single hole injectors.

Detailed Flame Measurements

Measurements reported by Michelfelder (3.31) illustrate the difference between three of the flame types descirbed in Section 3.2.1. Figure 3.5 shows the axial temperature distribution and flame length of four flames, two jet flames and two high intensity flames. The results illustrate:

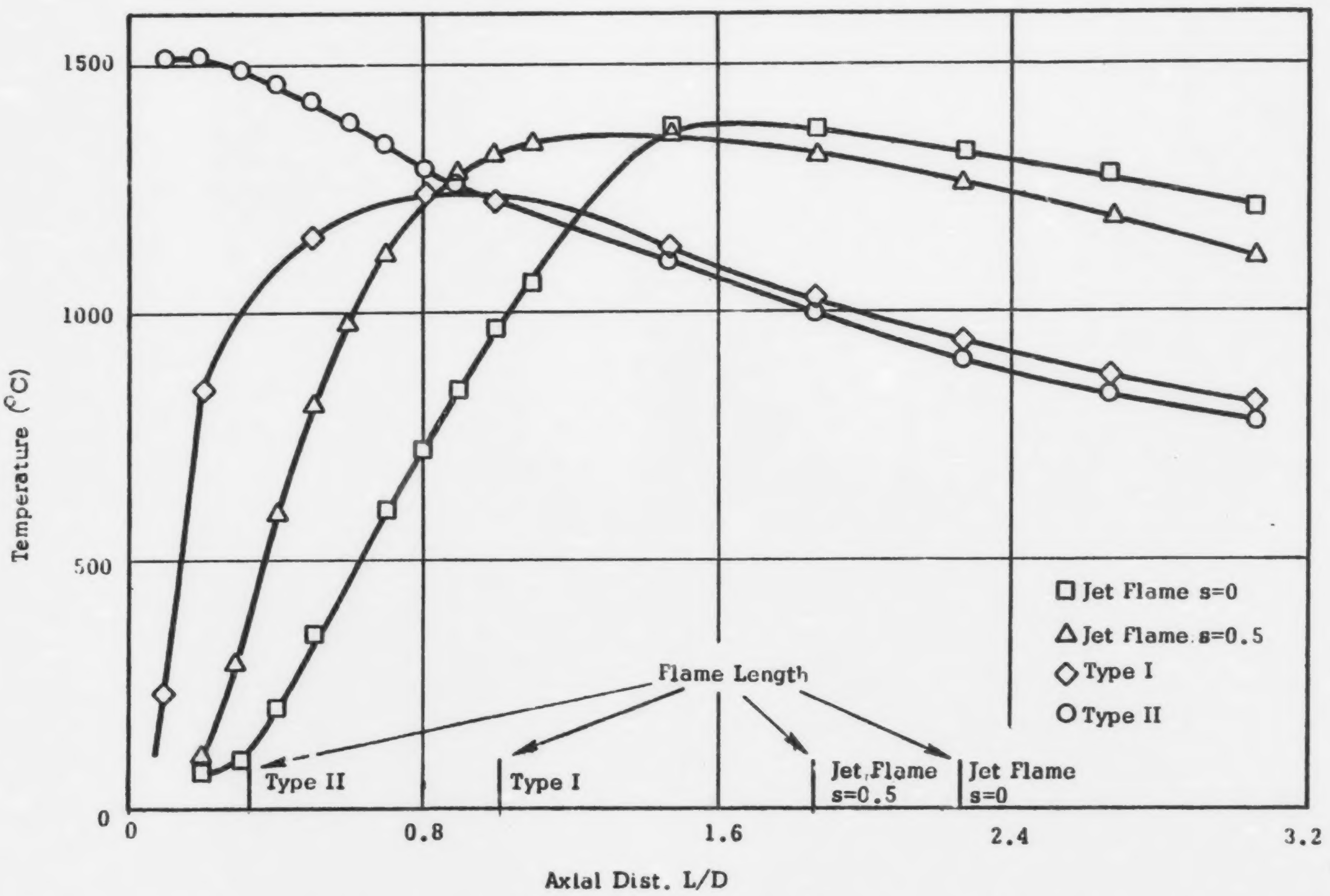


Figure 3.5 Axial Temperature Distribution Showing The Characteristics Of Different Flame Types (ref. 3.27)

- the application of weak swirl increases the rate of axial temperature rise in jet flames;
- the high initial temperatures in Type II flames are associated with the axial internal recirculation zone;
- Type II flames are shorter than Type I flames although the distribution of heat flux distributions from both flames is similar.

The flow patterns presented in Fig. 3.6 indicate the difference between the three high intensity swirling flames. This difference is also reflected in the gas concentration measurements shown in Fig. 3.7. Although the fuel jet remains on the axis with the Type III flame the methane concentration decays much more rapidly than in the Type I flame. This can be contrasted with the rapid reduction of methane concentration shown with the Type II flame.

3.2.2.2 The Characteristics of Swirling P.F. Flames

The increased rate of entrainment and mixing rates caused by the application of swirl to p.f. flames intensifies combustion and improves ignition stability. Since the rate of decay of axial velocity is increased by the application of swirl and coal burnout is time dependent, burnout distances are also reduced.

With a medium to high volatile coal and a single hole axial fuel injector at zero swirl a visible ignition front is established some distance downstream from the injector. This type of flame can be considered to be analogous to the lifted gas flame. The ignition distance is dependent on such factors as the:

- volatile content of the coal;
- primary to secondary velocity rates;
- degree of preheat;
- amount of primary air;
- temperature of the external recirculation;
- furnace wall temperature.

As the swirl is applied to the combustion air the ignition front moves upstream towards the burner. At some critical swirl level the ignition front jumps the

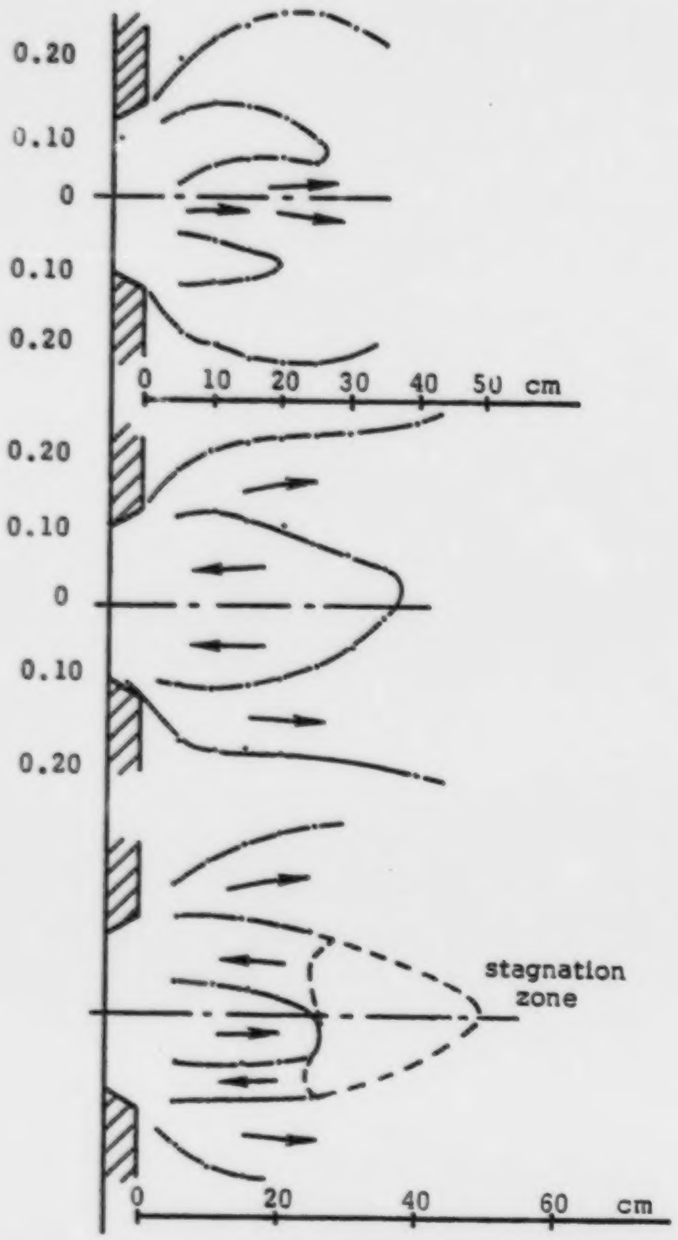


Fig. 3.6a
Type I

Fig. 3.6b
Type II

Fig. 3.6c
Type III

Figure 3.6 Flow Patterns Measured In Swirling Flames (ref. 3.27)

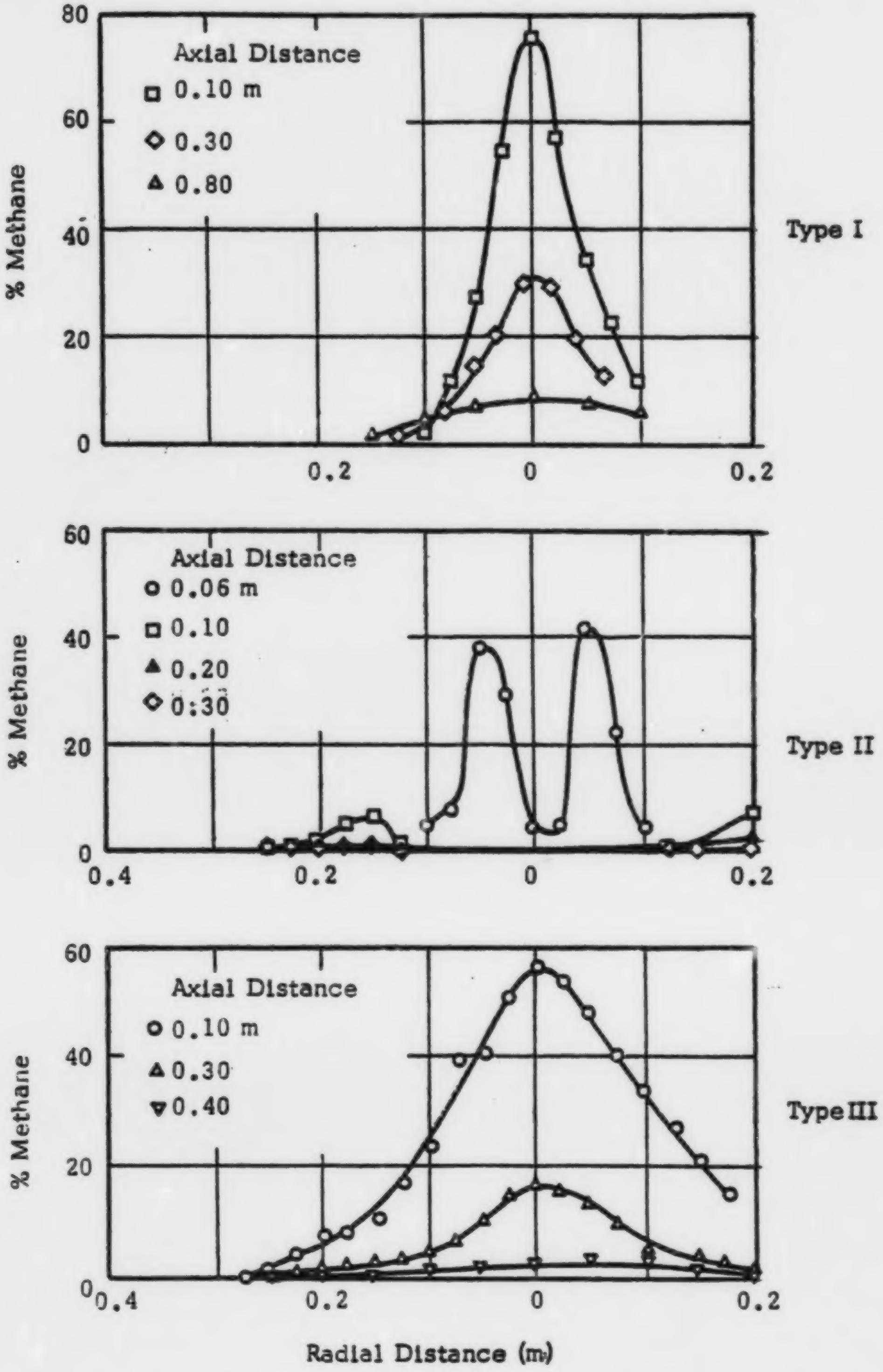


Figure 3.7 Radial Gas Concentration Measurements In High Intensity Flames (3.27)

distance to the injector. At this point, depending on the injector type, the four other flame types discussed in Section 3.1.1 may be formed:

- with low velocity axial or annular injectors a type II flame can be established initially;
- with slightly higher velocities or with increased swirl either a Type III or a Type I flame can be produced;
- with high velocity axial injector the flame looks like a simple jet flame even at high swirl levels when an annular reverse flow zone is present. This is because very little of the high momentum primary jet is entrained and returned to the base of the flame. Prior to the establishment of a stable ignition front the primary jet may become unstable. Under these conditions the coal jet is no longer directed along the axis but its direction varies randomly about the axis. In general the sequence of flame types with increasing swirl can be seen from the diagram shown in Fig. 3.3. It can be seen that with certain injectors continued increase in swirl after the formation of a Type II can allow a Type III flame to be formed. This phenomenon has not been reported with natural gas. It is believed that it is due to changes in the static pressure field associated with increased combustion intensity within the burner divergent.

The Influence of Burner Parameters on Ignition Stability and Burnout of P.F. Flames

The rate of combustion of p.f. is dependent on the temperature and composition of the gases surrounding the particles. Consequently, both ignition stability and burnout are related to the mixing pattern of the fuel jet, the combustion air and the recirculating gases. The heating rate of the coal particle will be dependent on the incident radiation both from the p.f. flame itself and from the furnace walls and upon convection from the surrounding gases. The burner parameters which influence the size of the recirculation zone and the mass of recirculating gases will influence the combustion characteristics of p.f. flames.

If low volatile coals are used then the axial temperature distribution provides a means of assessing ignition stability since a visible ignition front may not be present. The optimum condition for ignition is considered to be a steep axial temperature gradient. Figs. 3.8 and 3.9 indicate the influence of swirl on the axial temperature distribution and of anthracite burnout of flames.

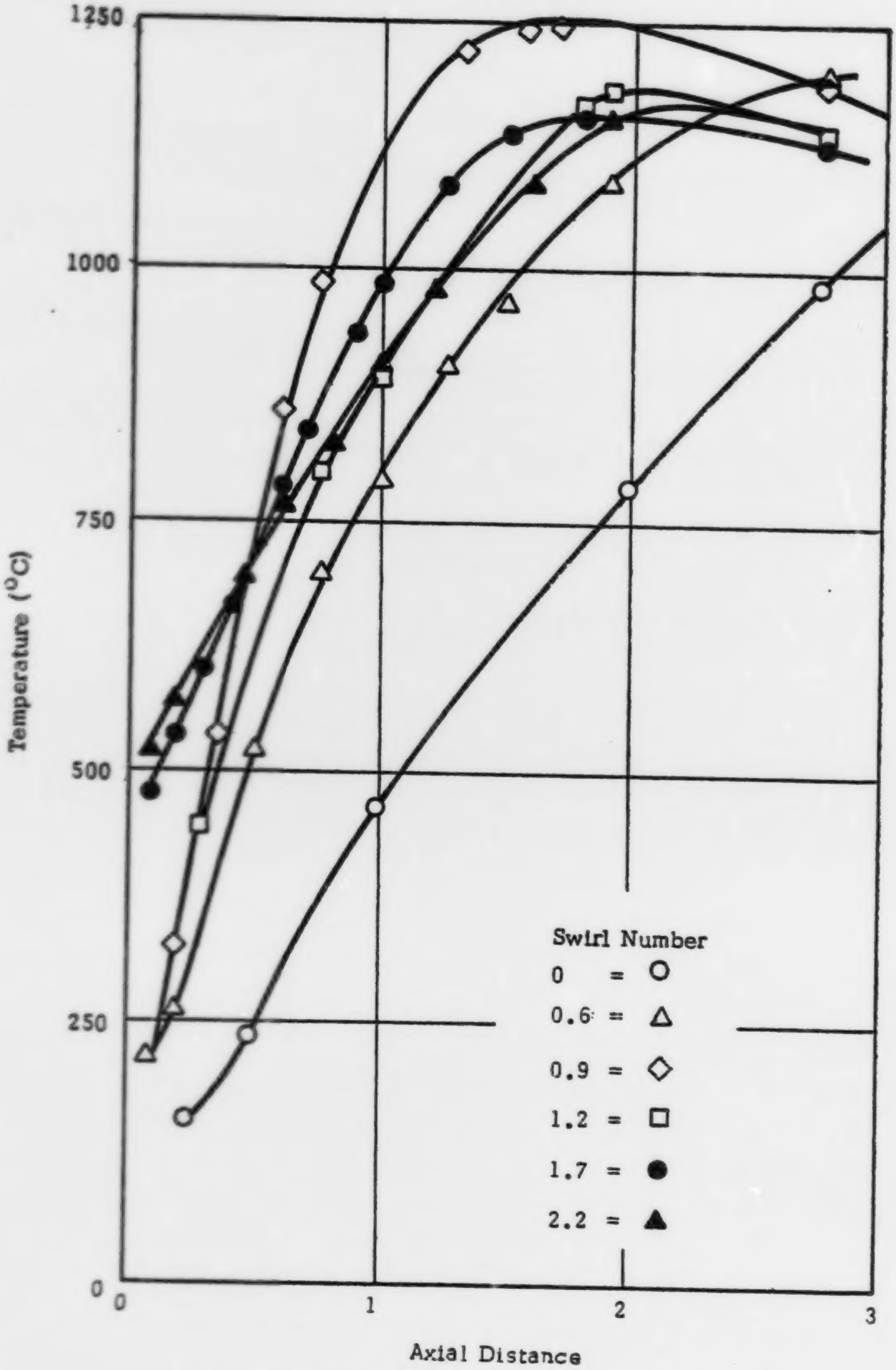


Figure 3.8 The Effect Of Swirl On The Axial Temperature Distribution Of Anthracite Flames (ref. 3.32)

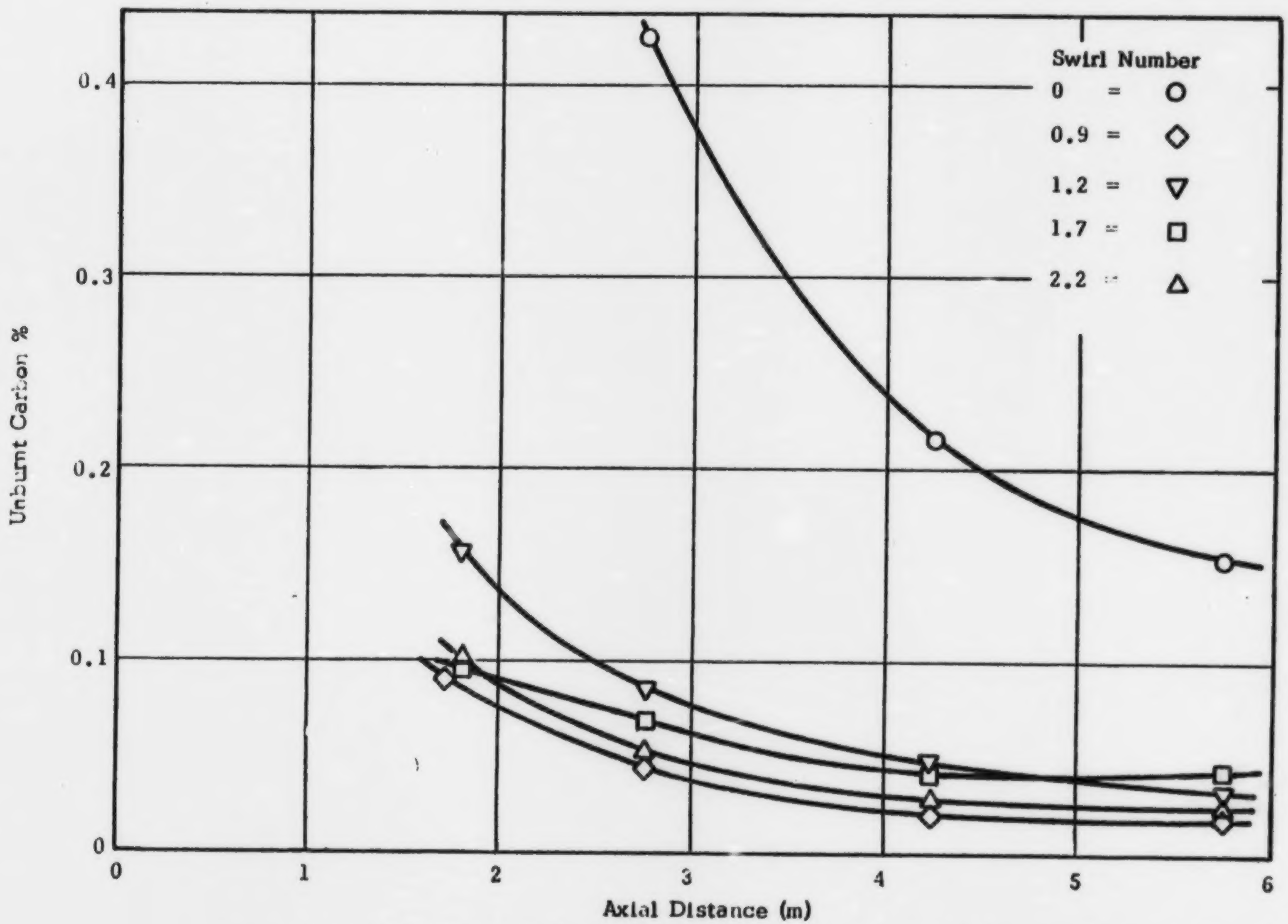


Figure 3.9 The Effect Of Swirl On The Burnout Characteristics Of Anthracite Flames (ref. 3.32)

It can be seen that there exists an optimum degree of swirl both for ignition stability and burnout. It can be seen from the radial temperature profiles (Fig. 3.10) that the critical condition is not a Type II flame. Although the base of the flame shows the characteristics Type I pattern the critical flame is more nearly classified as a Type III flame. The coal jet is not swept backwards by the recirculation zone but stagnates and then spreads some distance from the injection point. After the critical conditions have been attained any further increase in swirl causes a deterioration in the combustion characteristics. Hein (3.29)has shown that the optimum swirl level for early stable ignition of anthracite flames increases with increases in primary jet momentum and reduced secondary air velocity. The optimum swirl level is reduced if primary swirl is used, the burner exit is divergent rather than parallel and the length of the burner divergent is increased. It was possible to achieve visible ignition if 12% of the stoichiometric air requirement was used for the primary supply but not if 24% was used. Although visible igntion meant a steeper axial temperature rise it was not accompanied by reduced burnout distance. The fuel burnout is associated with residence time and this is controlled by the jet momentum and the swirl level.

Beer and Lee (3.30)have shown that the burnout of anthracite in swirling flames can be predicted by considering the furnace to be a combination of two adjacent reactors, one well-stirred and the other a plug flow reactor. By a combination of furnace measurements and isothermal experiments these workers were able to show that swirl influenced the mean residence time in the well stirred section.

Increasing the volatile content of the coal means that the ignition front will be visible and stable ignition can be considered to be achieved when this visible ignition front is stable at the injector. The visible ignition fron corresponds to the combustion of the volatile coal fractions which are released from the coal particles when they are heated. Thus, the conditions for stable ignition are:

- rapid heating of the coal particles to ensure an adequate quantity of combustible gas;

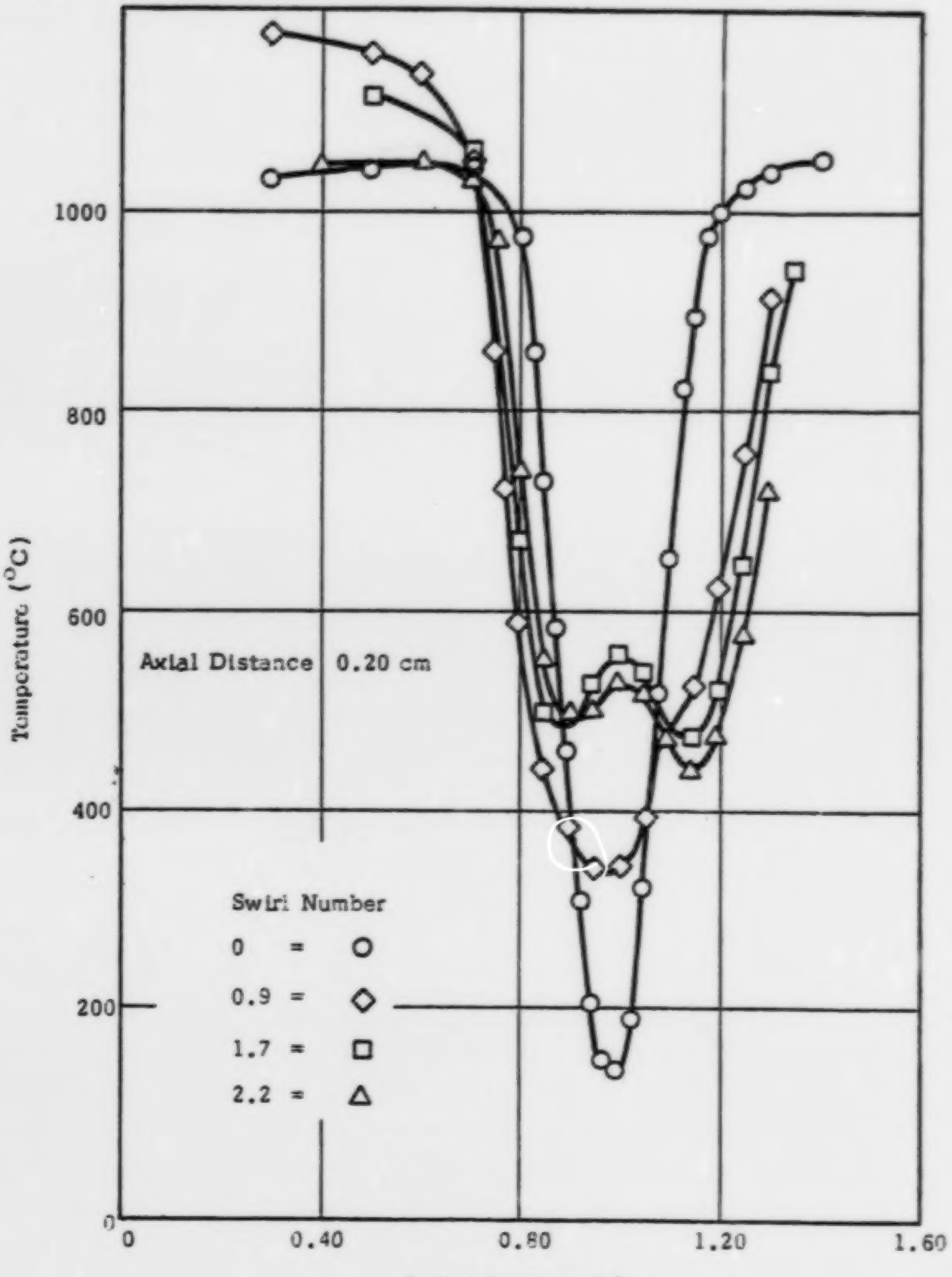


Figure 3.10 The Effect Of Swirl On The Radial Temperature Distributi Of Anthracite (ref. 3.32)

- the volatile gases must be mixed with air
- a stable ignition front must be present.

The satisfaction of these conditions necessitates rapid mixing both with air and with hot recirculating products (i.e., internal recirculation). Consequently, the burner parameters which control the size and strength of the recirculation zone will influence ignition stability. Thus divergent burner exits reduce the optimum swirl level from that necessary to ensure ignition stability with parallel exits. As with natural gas flames the method of fuel injection has a profound effect on stability. In this program it was possible to obtain stable ignition at zero swirl by the use of a radial coal injector from which the coal was injected though a series of holes around the primary pipe perpendicular to the burner axis.

As with low volatile coals burnout distances can be increased if the swirl level is increased beyond some critical level. This is illustrated by the results presented in Fig. 3.11.

The Influence of Coal Type on Flame Pattern

In the earlier discussions it was assumed that burner variables (swirl, geometry etc.) dictated the flow pattern (flame type, size of recirculation zone) in swirling flames. However, the temperature gradients associated with swirling flows also influence the flow pattern. Fricker and Leuckel (3.27) compared swirling isothermal flow with swirling natural gas flames and found:

- flow patterns existed in isothermal flow which were not found in swirling flames;
- phenomena (i.e., transformation from Type I to Type II) happended in isothermal flows at lower swirl numbers than in combusting flows.

The effect of combustion on the characteristics of swirling flows is also evident from the results presented in Fig 3.12 (ref. 3.32). These figures compare the flow patterns for identical burner conditions when burning two coals of different volatile contents. It can be seen that the flow pattern produced at medium swirl is Type I for the medium volatile coal and Type II for the low volatile fuel. At high swirl the internal recirculation zone is reduced in size when firing the medium volatile coal. Although measurements of static pressure were not made, it is believed

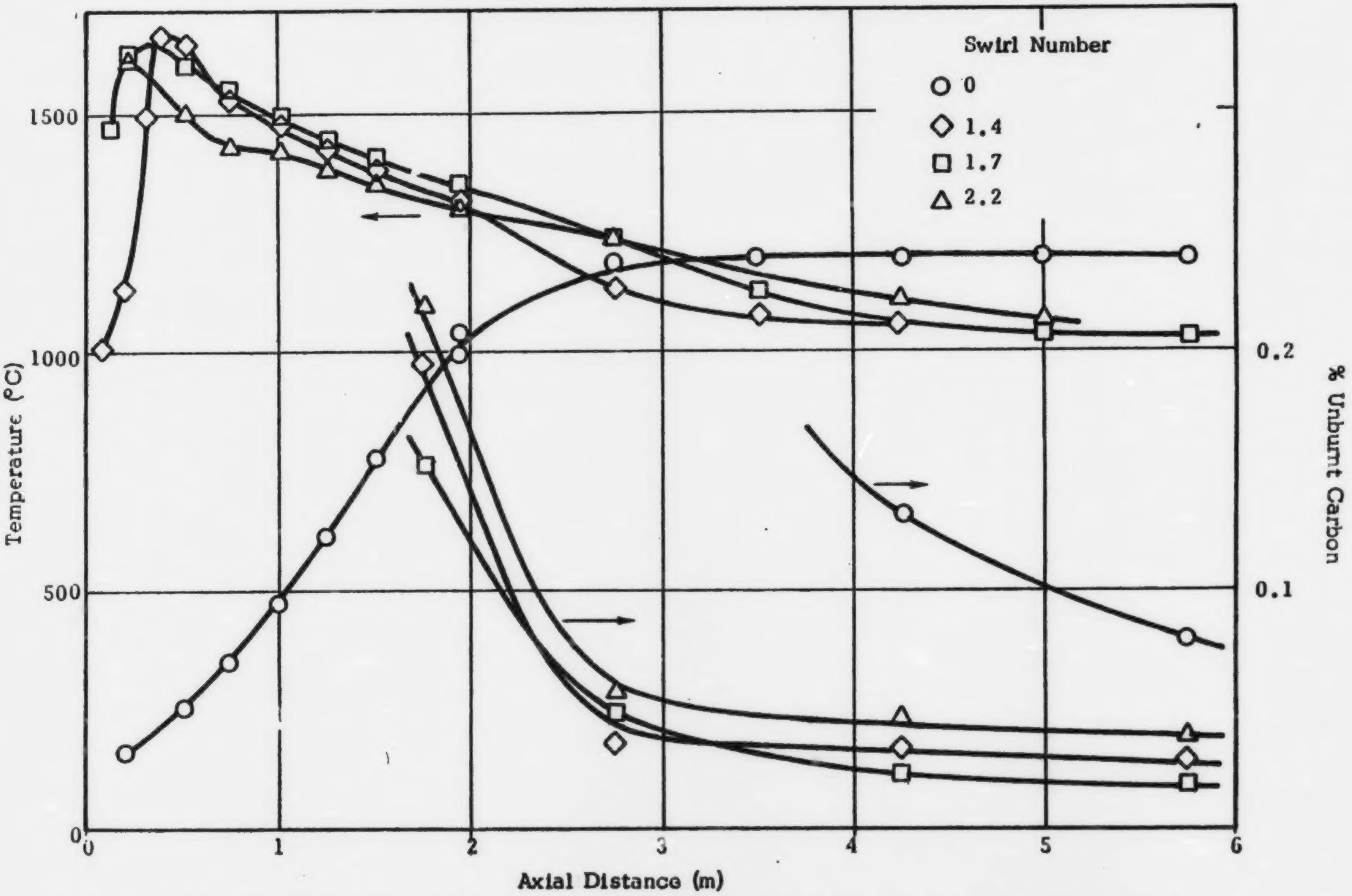


Figure 3.11 The Effect Of Swirl On The Axial Temperature Distribution And The Burnout Characteristics Of Medium Volatile Coal Flames (ref. 3.32)

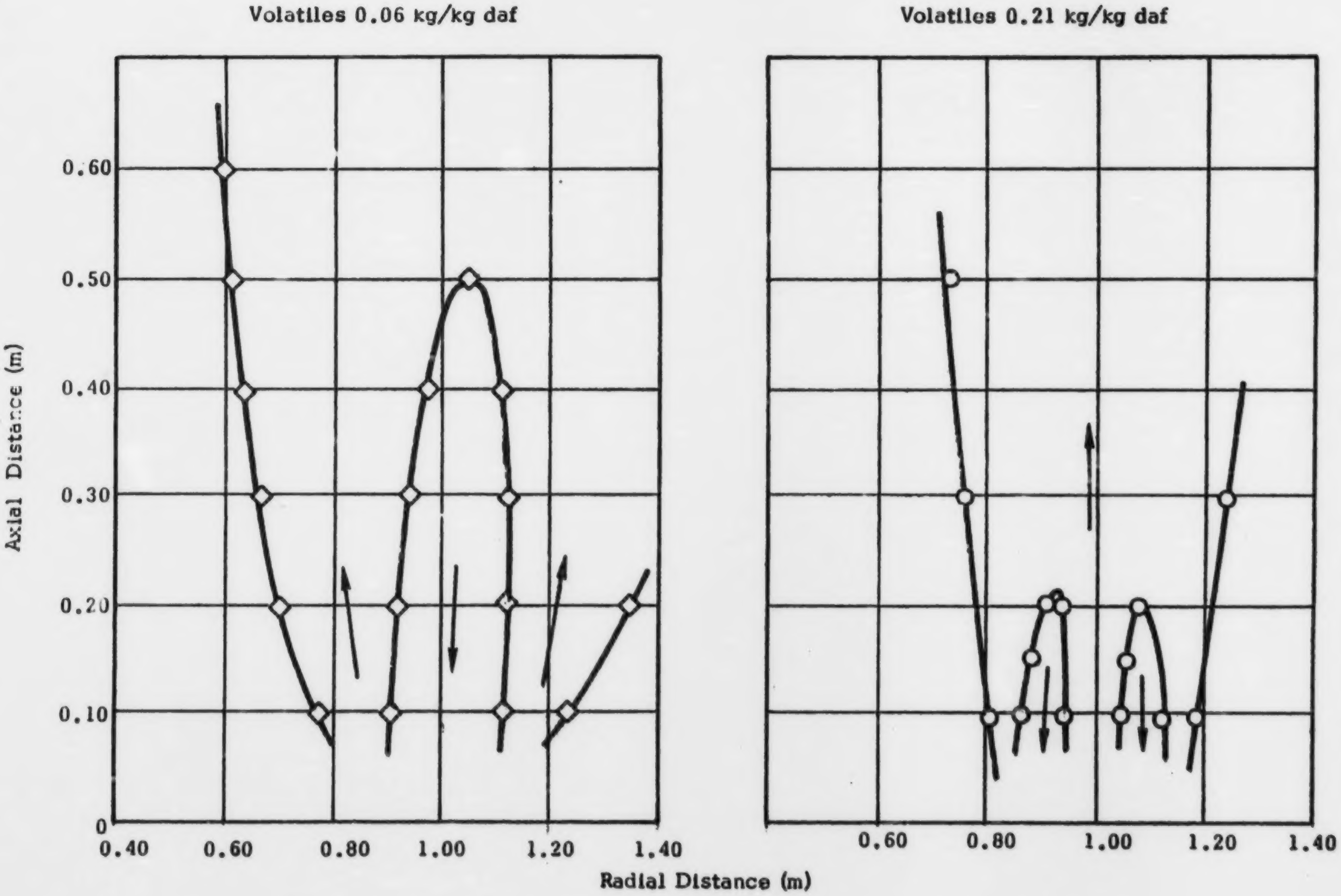


Figure 3.12 Influence Of The Volatiles On The Flowpattern (Swirl No. 1.7)

that the static pressure field is influenced by the higher temperatures and steep temperature gradients produced by the combusting volatile coal fractions.

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4. TEST EQUIPMENT AND OPERATIONS

The experimental results presented in this report were obtained in two separate trial series:

- M-2 trials (intermittent measurements during Oct. Nov. Dec. 1971), evaluation of measuring equipment for gas flames and an investigation of the effect of furnace cooling load on the emission of nitrogen oxides from gas flames;
- AP-1 trials (continuous furnace operation from March 17th to May 16th, 1972), an investigation of the effect of burner parameters on nitrogen oxide emissions from natural gas and p.f. flames.

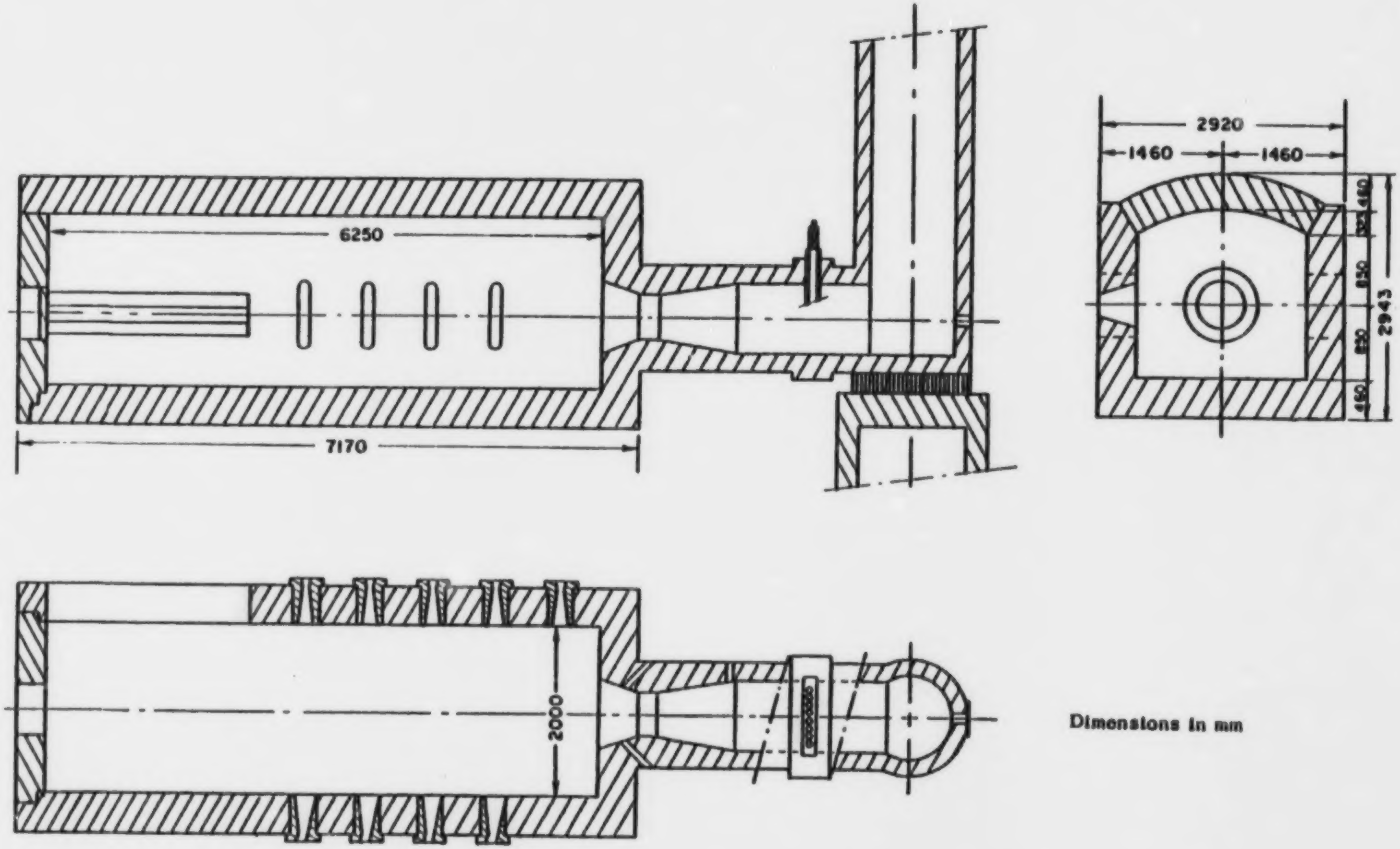
The AP-1 trials can be subdivided into four separate sections:

- natural gas I/O;
- p.f. I/O with low nitrogen coal;
- flame mapping;
- p.f. I/O with high nitrogen coal.

The furnace was operated continuously during the trial period. The measurements necessary to enable complete velocity, temperature and concentration maps of the flame to be drawn were made by four, two man teams working 3 shifts per 24 hours. No fixed shift system was used for the I/O investigations; the measuring times were dictated by the necessity for allowing furnace conditions to equilibrate after changing burner parameters. Details of the furnace, input conditions and burner design used during the investigations are presented in this section.

4.1 Furnace Conditions and Inputs

The furnace investigations were carried out in the Ijmuiden No. 1 furnace a plan of which is presented in Fig. 4.1a. The furnace is a horizontal, refractory tunnel of approximately square cross section, with internal dimensions 2m x 2m x 6.25 m. In the M-2 trials the furnace was cooled by a symetric arrangement of seventeen cooling loops (illustrated in Fig. 4.1b). However, for the AP-1 trials, the cooling system was removed and the furnace was operated virtually uncooled. In order to prevent the furnace stack from overheating



Dimensions in mm

Figure 4.1a Experimental Furnace No. 1 As Used For The AP Trials

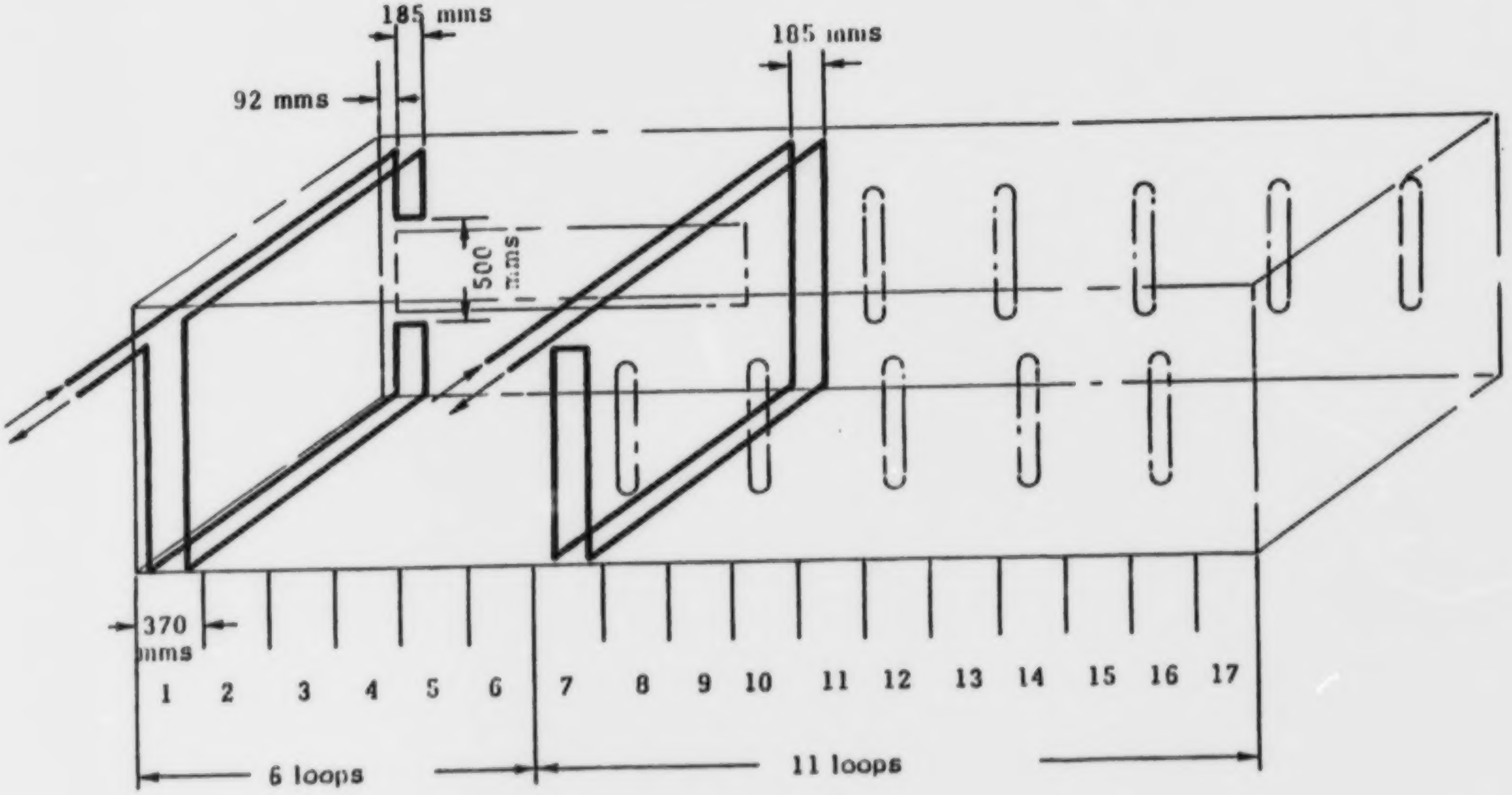


Figure 4.1b Schematic Arrangement of Cooling Pipes In Furnace No. 1

16 cooling pipes were placed in the final 2 m of the furnace length; the pipes were evenly divided between the furnace roof and hearth. Access to the furnace for both measurement and observation was provided by the horizontal and vertical slots (see Fig. 4.1). The horizontal slot enabled continuous probe movement in an axial direction for a distance of 2m from the furnace wall. In leakage of air was prevented by operating the furnace at a slight overpressure and using close fitting doors for the measuring slits.

The Slochteren natural gas used during the investigation had the following nominal composition:

CH_4 - 81.3% v/v
C_2H_6 - 2.9% v/v
C_3H_8 - 0.4% v/v
C_4H_{10} - 0.1% v/v
C_nH_m - 0.1% v/v
CO_2 - 0.8% v/v
N_2 - 14.4% v/v

The firing rates used during the investigation were:

AP-1 200 Nm^3/hr (heat input of 1.76 Mw)
M-2 340 Nm^3/hr (heat input of 3.00 Mw)

Two coals of different nitrogen contents were used in the AP-1 trials. The majority of the work was carried out with a low nitrogen coal from Lorraine, France which had the following properties:

- volatile content 32.7% (on a dry basis)
- ash content 6.3% (on a dry basis)
- carbon content 78.48% (on a dry basis)
- hydrogen content 4.77% (on a dry basis)
- nitrogen content 1.05% (on a dry basis)
- sulphur content .75% (on a dry basis)

A firing rate of 204 kg/hr of coal was used which assumed a moisture content of 2% to give an equivalent thermal input to 200 Nm^3 of natural gas i.e., 1.76 Mw.

Experiments were also carried out with a high volatile (35%) coal from Northumberland, England with a nitrogen content of 1.8%.

4.2 Burner Design

The burner used for the investigations is shown in Fig. 4.2 and was positioned to fire along the length of the furnace from the center of the front wall. The burner can be described as a double concentric jet burner with variable swirl intensity in the annular air supply. The burner was not designed to represent any particular practical burner but it is typical of all double concentric systems. One of the features of the burner was its versatility; it readily allowed the following parameters to be varied:

- the type of fuel injector;
- the position of the fuel injector relative to the burner exit. Although the design allowed continuous axial movement, only the throat and axial positions were investigated;
- the velocity of the combustion air;
- the angle of the burner exit;
- the type of burner divergent (refractory or water cooled);
- the degree of rotation (swirl) in the annular combustion air stream.

The swirl was produced by a moving block swirl generator which is shown in Fig. 4.2. The swirl register consists of two annular plates (P_1 and P_2) and two series of interlocking wedge shaped blocks (B_1 and B_2) each attached to one of the plates, interlocked the blocks form alternating radial and tangential air flow channels, such that the air flow splits into an equal number of radial and tangential streams which combine downstream from the actual swirl generator into one swirling flow. Rotating the back plate P_2 (the plate P_1 is fixed to the burner) progressively closes the radial channels and opens the tangential ones (or vice versa). Thus the resulting flux of angular momentum increases (or decreases) continuously between zero and a maximum value dependent upon the dimensions of the system.

Four different types of fuel injector were used during the investigations and they are classified with reference to the type of injection orifice:

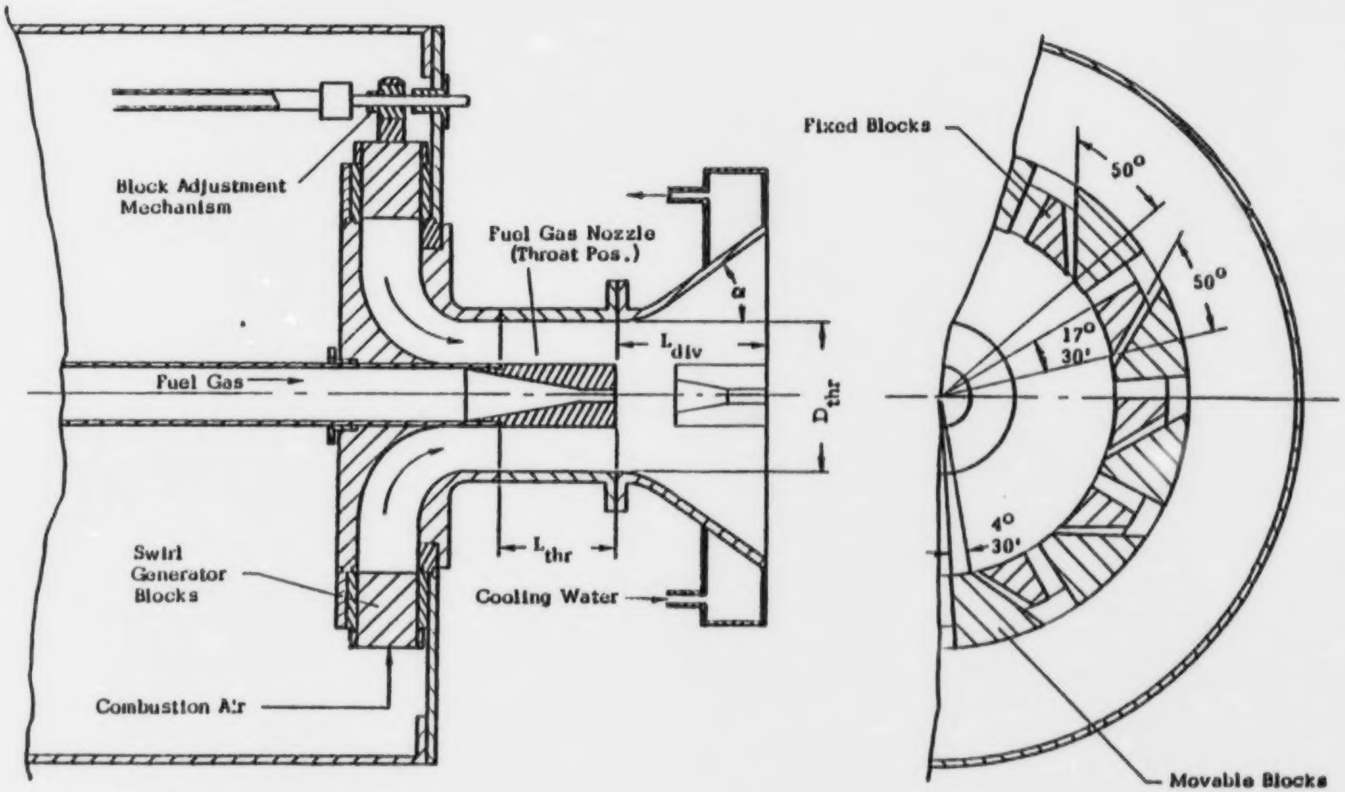


Figure 4.2 Double Concentric Burner System

- single hole;
- multihole divergent;
- multihole radial;
- annular.

The dimensions of the various injectors are given in Table 4.1.

Table 4.1 GAS INJECTORS

Nozzle number	Nozzle type	Length of hole mm	Diameter of hole mm	Gas exit Velocity m.sec^{-1}
1	single hole	99.2	46.1	35
8	single hole	26.0	13.0	383
19	16 divergent holes at 35°	-	4.7	200
32	16 radial holes		9.65	50
33	16 radial holes		4.7	200

5. MEASUREMENT SYSTEMS

The amount of nitrogen oxides emitted from fossil fuel fired furnaces depends upon the detailed time-temperature-concentration history of the input fuel and air and the products of combustion. During the AP-1 trials the following properties were measured:

- temperature;
- velocity;
- gas and solid concentrations;
- nitrogen oxide concentrations.

In order to determine the spatial distribution of nitrogen oxide within flames it was necessary to develop new sampling systems. The other properties were determined by measuring systems developed by the I.F.R.F. and described by Chedaille and Braud (5.1).

5.1 Temperature Measurement

Suction pyrometers were used to measure temperature since these give an accurate direct reading of temperature. The temperature is measured by a thermocouple positioned on the axis of a system of shields and the gas whose temperature is to be measured is operated through the annular channels formed by the shields (see fig. 5.1). The shields serve a dual purpose:

- they isolate the thermocouple from the surrounding radiation;
- they protect the thermocouple from contamination by the furnace matter.

The efficiency of the pyrometer increases as the suction rate increases up to velocities of the order of 250 $msec^{-1}$.

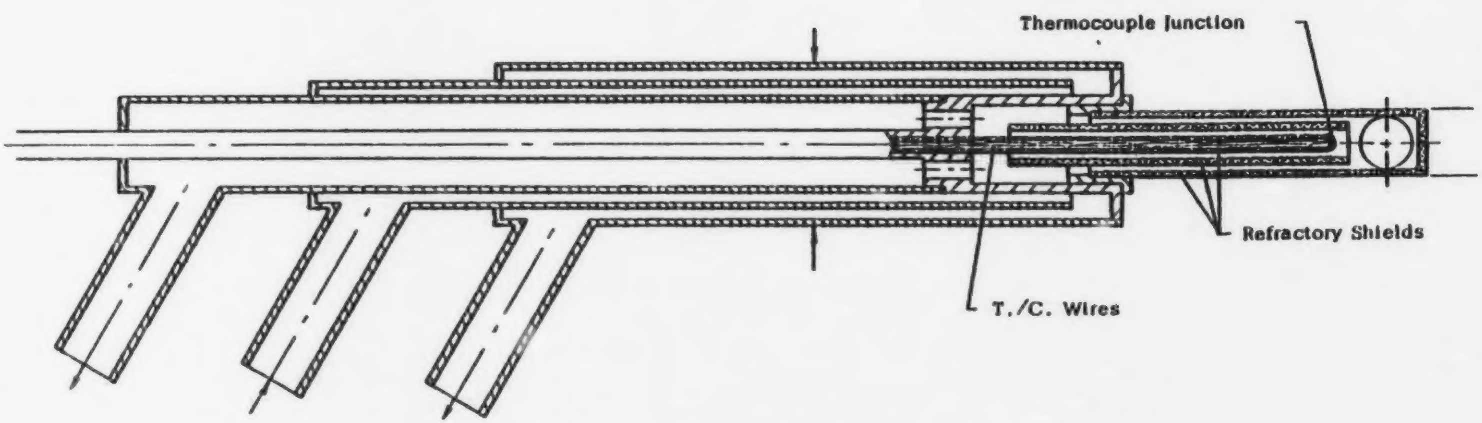


Fig. 5.1 Suction Pyrometer with Replaceable Refractory Shields

During the measurement period the suction mass flow rate was continuously monitored by an orifice plate and suction velocities greater than 200 $msec^{-1}$ were maintained. The solids in p.f. flames tend to block the shields and necessitate frequent shield changes even though the wake sampling position (Figure 5.1) is used with p.f. flames.

The construction of the pyrometer almost eliminates many of the errors associated with bare thermocouples. However, the measured value may be in error because:

- the sampled matter may contain solid particles whose temperature is different from that of the gas and the measured temperature will be neither gas nor the particle temperature;
- within the "flame" the sample will contain fuel and oxygen which are not mixed on the molecular scale and combustion will not take place. It is possible that micromixing could be intensified at the high suction velocities causing excessive after-combustion within the shields.
- the measured temperature is some time averaged value of a fluctuating temperature. This temperature may be biased because of the effect of viscosity changes on sample flow.
- spatial resolution. The zone of high temperature may be small compared to the sample volume.

5.2 Velocity Measurements

Detailed measurements were made in highly swirling flames during the AP-2 trials where the three-dimensional nature of the flow requires both the magnitude and direction of the flow to be known. It is not possible to use standard yaw and pitch meters because rotation of the probe is inconvenient under furnace conditions. Consequently, measurements of velocity magnitude and direction were made with a hemispherical five-hole pitot probe. These properties are determined from the measured pressure distribution around the head of the probe when placed in the flowing stream.

Lee and Ash (5.2) have demonstrated that with five holes on the circumference of a sphere it is possible to calibrate an instrument to enable the magnitude and direction of the flow in three dimensional field to be determined. Details of the probes used in the AP-1 trials are shown in Fig. 5.2. Before and after use the probes are calibrated in a wind tunnel. The calibration procedure developed by Leuckel (5.3) allows the rapid evaluation of the desired properties from measurements of five pressure differentials: $P_o - P_3$, $P_1 - P_3$, $P_o - P_4$, $P_2 - P_4$ and P_o - Patm.

The static pressures were recorded by mutual inductance transducers with a sensitivity of .01 mm water column and electronically integrated. Measurements were made with the probe pointing in the direction of mean axial flow. In the boundary regions measurements were made with the probe directed both towards and away from the burner and the required velocity profile was obtained from the combined measurements. Decisions concerning the flow direction (either towards or away from the burner) were based upon the measurements themselves and checked by referring to measurements made by a two-hole cylindrical probe.

Errors involved in measurements with impact tubes have been adequately discussed elsewhere (5.1). Measurements in pulverized fuel flames introduce further complications due to the presence of solid particles. In gas and oil flames solid concentrations rarely exceed a few milligrams per litre and mean diameter is normally less than 1μ. In p.f. flames particle sizes range from 1μ to 200μ and concentrations are of the order of several grams per litre. Consequently, coal particles affect the velocity measurements by virtue of:

- deposition upon the probe surface. Large scale deposition upon the head will affect the calibration of the probe;
- blockage of the pressure holes, which will result in damping of the fluctuations and a drift in the signal;
- variation in fluid density. Both the static and total pressure heads will depend upon the concentration and size of the particles. The velocities were calculated assuming the fluid density to be given by

  $$\rho_{fluid} = \rho_{gas} + C$$

  where C is the particle concentration under the measuring conditions.

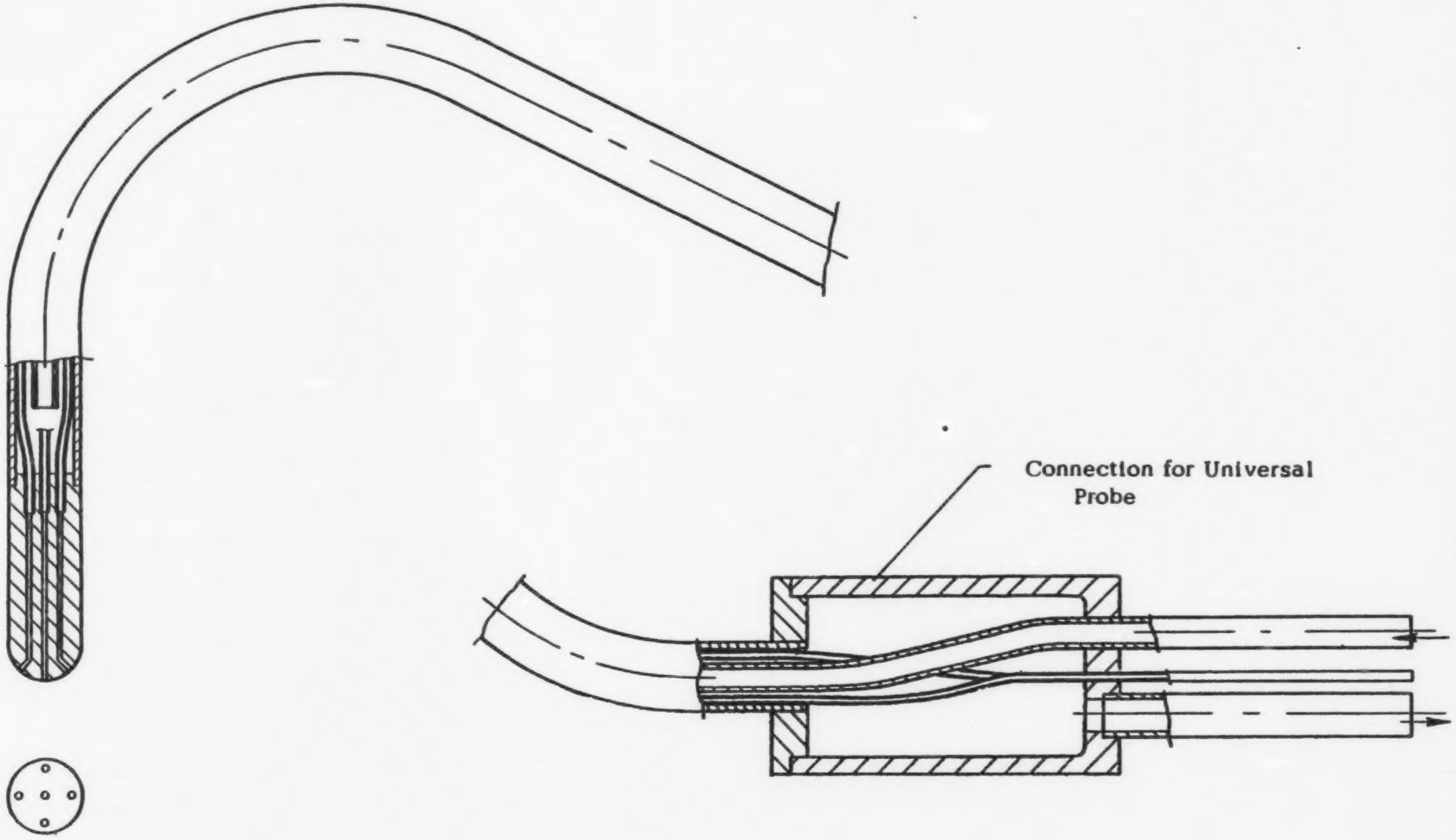


Fig. 5.2 5-Hole Impact Probe

The problems associated with probe blockage and particle deposition cannot be eliminated. However, it has been found that problems can be reduced if the head is not completely cooled. Thus, the head is heated by radiation and any deposits burn away. A disadvantage of this system is that the combustion of particles either on the surface or in the pressure holes will also produce errors. Pressure hole blockage can also be reduced by frequent purging with compressed air.

Even when precautions are taken velocity measurements in high volatile coal flames are extremely difficult. The particles are sticky and measurements of the complete field are impossible using a five hole probe.

5.3 Species Concentration Measurements

The measurements of species concentration in flame gases involves two separate operations:

- the collection and delivery of a representative sample to the analytical equipment;
- the analyses of the sample.

During the AP-1 trials the concentration of following species were determined:

- gases (N_2, O_2, CO_2, CO, H_2, CH_4, C_2H_6, C_3H_8, C_2H_4, C_nH_m, NO, NO_2);
- solids.

The analytical equipment is listed in Table 5.1. Both the chromatograph and chemiluminescent analyzer were constructed from component parts. The chromatograph allows the concentration of specific hydrocarbons to be identified and quantified. Automatic operation with a program sequence of three detectors and five columns allows the analysis of the major components to be completed in thre

TABLE 5.1 ANALYTICAL EQUIPMENT

Class	Species	Analytical equipment	
Flue gases	O_2	Paramagnetic	
	CO	Infrared	
	CO_2	Infrared	
Flame gases		Gas Chromatograph	
		Carrier gas	Column material
	H_2	Nitrogen	Molecular sieve
	CO_2	Helium	Silica gel
	O_2, N_2, CH_4 CO	Helium	Molecular sieve
	Hydrocarbons	Helium	Alumina
	NO	Chemiluminescent analyser	
	NO_2	Chemiluminescent analyser with quartz converter	
Solids	Ash, fixed carbon, volatiles	Pozetto	
	Nitrogen	Coleman	

minutes. The chemiluminescent analyzer was built to E.P.A. specification and operated satisfactory in the NO mode. NO_x concentrations were determined by diverting the sample through a heated packed quartz tube to convert nitrogen dioxide to nitric oxide.

Stainless steel was not used because it is known that NO can be reduced if passed through heated steel tubes in reducing mixtures (5.4). Although the converter operated satisfactorily with completely combusted products NO_x concentrations were less than the NO values when the sampled gases contained H_2, CO and hydrocarbons. Consequently, NO_2 concentrations could not be determined in the flame gases.

The function of any sampling system is to collect a representative specimen and ensure that the sample does not undergo further reaction after collection. In order to satisfy the former requirement it is advisable to use a continuous sampling system. In this way it is possible to take account of the concentration fluctuactions which are inevitable large turbulent difussion flames.

The latter requirement is less easily satisfied when sampling gases containing nitrogen oxides. The nature and concentration of the oxides of nitrogen necessitated the development of a separate sampling system for nitrogen oxides from that used for the other species.

5.3.1 Sampling Systems for Nitrogen Oxides

The sample which is withdrawn from the furnace is at a high temperature and normally in a highly reactive state. Rapid cooling of the sample (to less than 300°C in less than 3×10^{-3} seconds) effectively quenches the reactions involving permanent gases, p.f. particles or soot (5.1). However, several workers (5.4 and 5.5) have reported that under certain circumstances normal sample probe materials, particularly stainless steel are not inert with respect to nitrogen oxides. Although nitrogen oxide measurements have been made in p.f. flames prior to this investigation (5.6), these workers did not indicate whether checks were made to ensure that reactions involving char particles and nitrogen oxides did not cccur within the sampling equipment. Consequently experiments were carried out to establish design criteria for probes capable of sampling nitrogen oxides from coal flames.

The most comprehensive test of any sampling system is the measurement of known concentrations under representative operating conditions. Such a test is difficult to carry out when the measurement concerns the NO content of flame gases, whose time mean temperature may exceed 1700°C and contain coal particles in a state of rapid decomposition. Therefore, it was decided to determine under which conditions certain materials were no longer inert with respect to nitrogen oxides by passing selected mixtures through heated tubes of the material to be tested. The tubes were heated in an electric furnace whose temperature could be controlled up to 1200°C. The nitric oxide content of the gas mixture was determined by a chemiluminescent analyzer before and after passing through the tube under test.

Three tube materials were tested:

- stainless steel;
- gold plated stainless steel;
- quartz;

and the results confirmed the conclusions of Halstead (5.4) and Shaw (5.5). Quartz is the only suitable material for sampling from reducing gases. A summary of the test results are presented in Table 5.2a. Although a probe constructed from stainless steel should be satisfactory in oxidizing conditions it was decided that a probe constructed with a quartz sampling tube would be used in natural gas flames. A diagram showing the construction of the probe is presented in Fig. 5.3. The quartz tube was ground to ensure good contact with the water cooled stainless steel liner. The gases are rapidly cooled to less than 2°C in a gas cooler at the probe exit ot remove water vapour.

Sampling from p.f. flames involves the separation of the solid and gaseous phases at some stage. This is normally accomplished by filtration close to the sample point to prevent blockage of the sample lines. Further experiments were

Table 5.2a TEST TO DETERMINE SUITABILITY OF MATERIALS FOR SAMPLING PROBES

Material	Gas Mixture	Temp. Range °C	Effect
Quartz	A	20-1000°C	None
Quartz	B	20-1000°C	None
Gold Plated s.s.	A	10- 720°C	None
Gold plated s.s.	A	725°C	4% loss of NO
Stainless steel	A	20- 590°C	None
Stainless steel	A	595°C	60% loss of NO

Table 5.2b EFFECT OF TEMPERATURE

Temperature Range	Mixture B Dusted Tube	Mixture B, Coal Dust Supported By Quartz Wool Plug	Mixture C, Coal Dust Supported By Quartz Wool Plug
70-170°C		no effect	
20-240°C			no effect
20-840°C	no effect		
170-250°C		8% increase of NO	
500		80% loss of NO	
620		100% loss of NO	70% loss of NO
1000	10% loss of NO		
1100	20% loss of NO		
1170	25% loss of NO		

Table 5.2c GAS MIXTURES USED IN PROBE MATERIAL TESTS

Constituent	Mixture A	Mixture B	Mixture C
H_2		8.5%	
CO_2		2.9%	
CH_4		5 %	
CO	36%	6 %	
C_2H_6		0.2%	
C_2H_4		0.3%	
C_3H_8		0.2%	
N_2	64%	66.9%	100%
NO	80 ppm	80 ppm	121 ppm

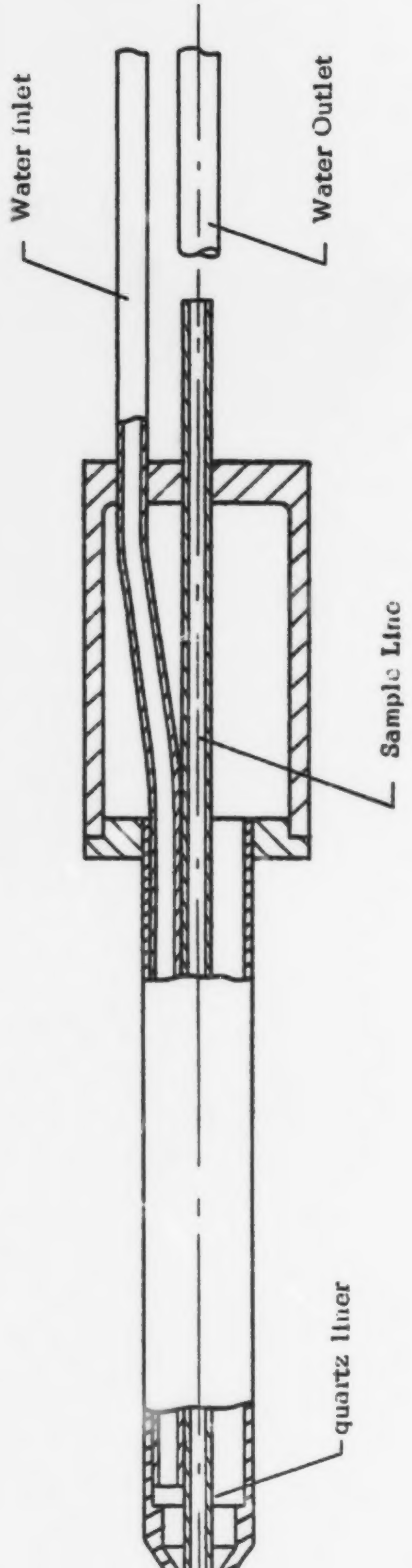


Figure 5.3 Water Cooled Quartz Lined Probe for Sampling From Gas Flames

carried out to assess the effect of char particles on the sampled gases containing nitrogen oxides. Char was supported on a quartz wool plug held in a quartz tube.

1. The effect of cold coal particles

   During this series of tests the gas mixture leaving the heated tube was passed through a "plug" of cold pulverized coal supported on a quartz wool filter. The tests described in Table 5.2a were repeated with the same results.

2. The effect of heat char particles

   The intert nature of quartz had already been demonstrated and these tests were restricted to the effect of hot coal particles in quartz tubes. In the first instance the coal particles were dusted into the tube surface before it was placed in the furnace and in the second test the coal was supported on a quartz wool plug. Gas mixture C was also passed through a plug of coal. Results of these tests are shown in Table 5.2b.

The results have important implications for the construction of probes suitable for sampling from p.f. flames since they show that contact between the gas and hot char particles cannot be tolerated.

Three probes were designed and built for testing in p.f. flames:

- a filter probe (Fig. 5.4);
- a water quench probe (Fig. 5.5);
- a steam quench probe (Fig. 5.6);

The probes were designed to ensure that the following conditions were satisfied:

- that any metal surface which was in contact with the sampled gases was adequately cooled;
- that the solid particles were rapidly quenched;
- that the probe was simple to operate and capable of withstanding flame conditions (high temperatures and solid loadings.)

The three probes were tested in both gas and p.f. flames and the steam quench probe was found to be unsatisfactory; the probe was blocked before a representative sample could be collected.

Both the water quench probe and the filter probe could be used in p.f. flames and similar concentrations were determined with both probes. These two probes were also compared with the quartz probes in a gas flame. The recorded concentrations are shown in Fig. 5.7. and it can be seen that there is no significant difference between the values obtained with the different probe types.

The water quench probe had one considerable advantage over the filter probe. It could be used continuously for long periods without blocking. The filter probe was cooled so efficiently that condensation occurred in the filter housing and the filter was rapidly blocked even with low solid loadings. Therefore the water quench probe was used to sample from p.f. flames.

5.3.2 Sampling Systems for Gaseous and Solid Material

The flame conditions dictated the type of probe which was used to collect flame gas and solid samples. Two types of probes with centered bronze filters to collect solid matter were used in p.f. flames (see Figs. 5.8 and 5.9). The two designs of filter probes were necessary because in regions of high solids loadings (e.g., the initial stages of p.f. flames) the small filter blocks rapidly and an accurate determination of the gas volume is not possible. The

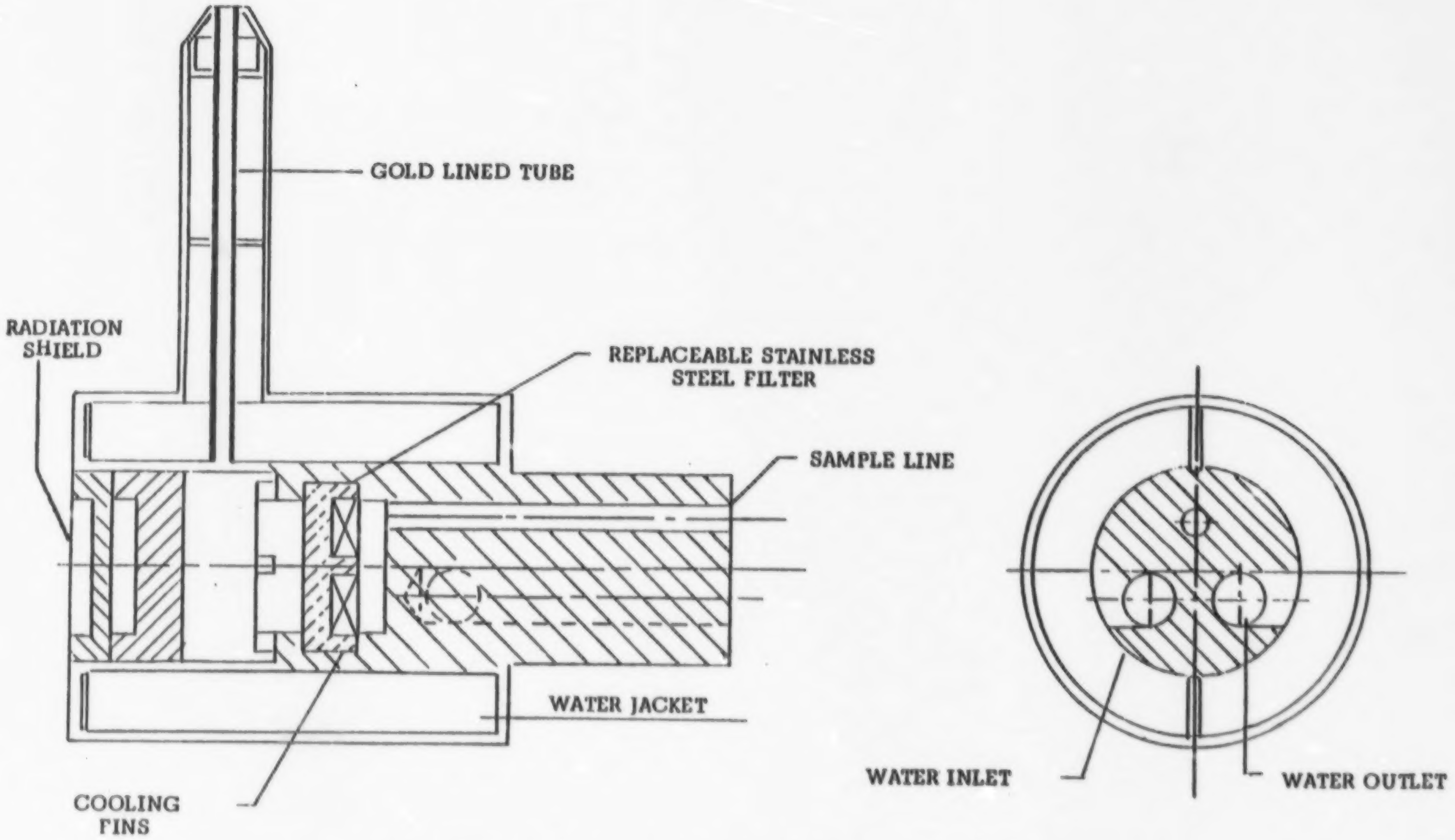


Figure 5.4 A Heavily Cooled Filter Probe For Use In P.F. Flames

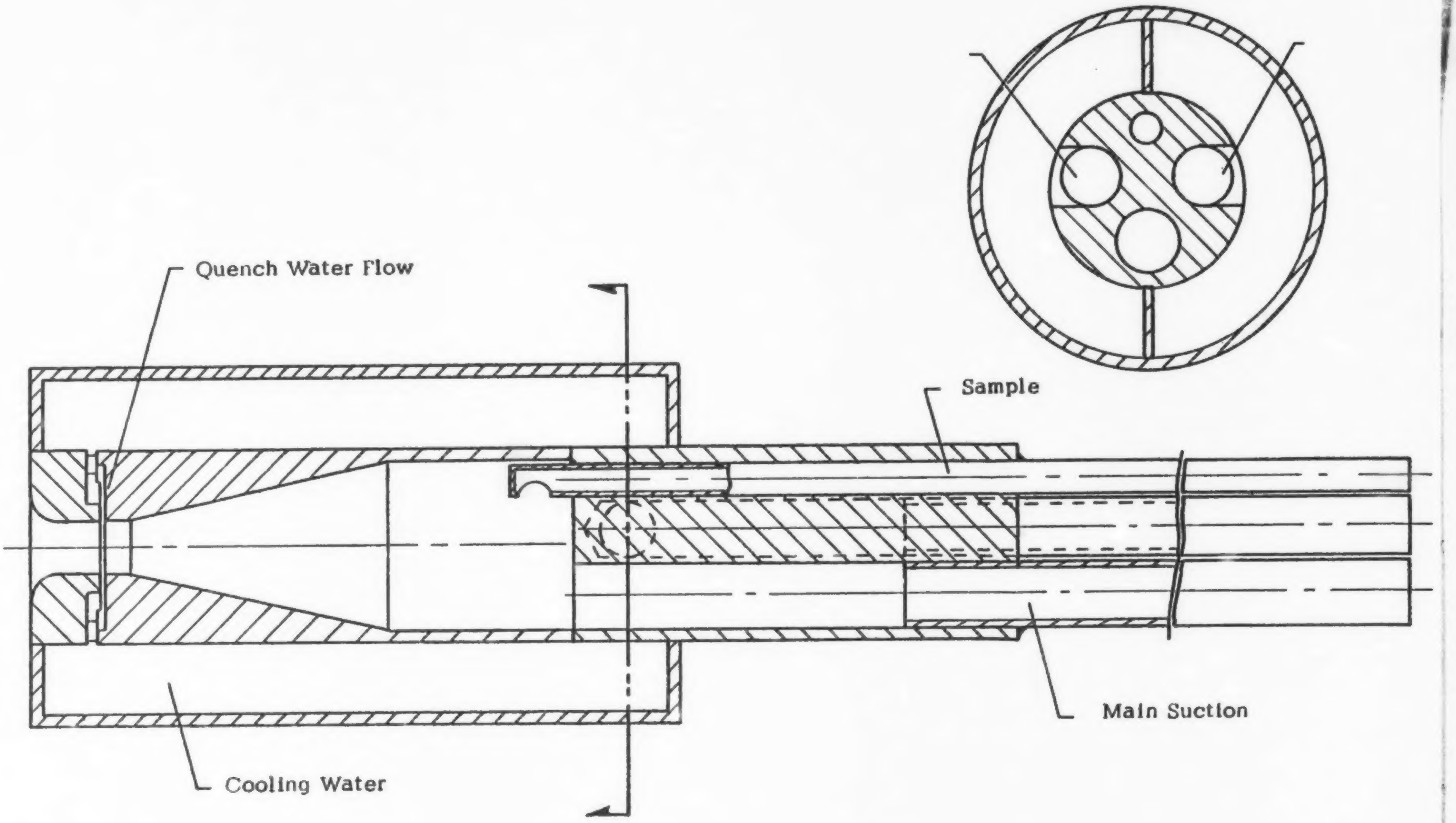


Fig. 5.5 Water Quench Probe for NO Measurement

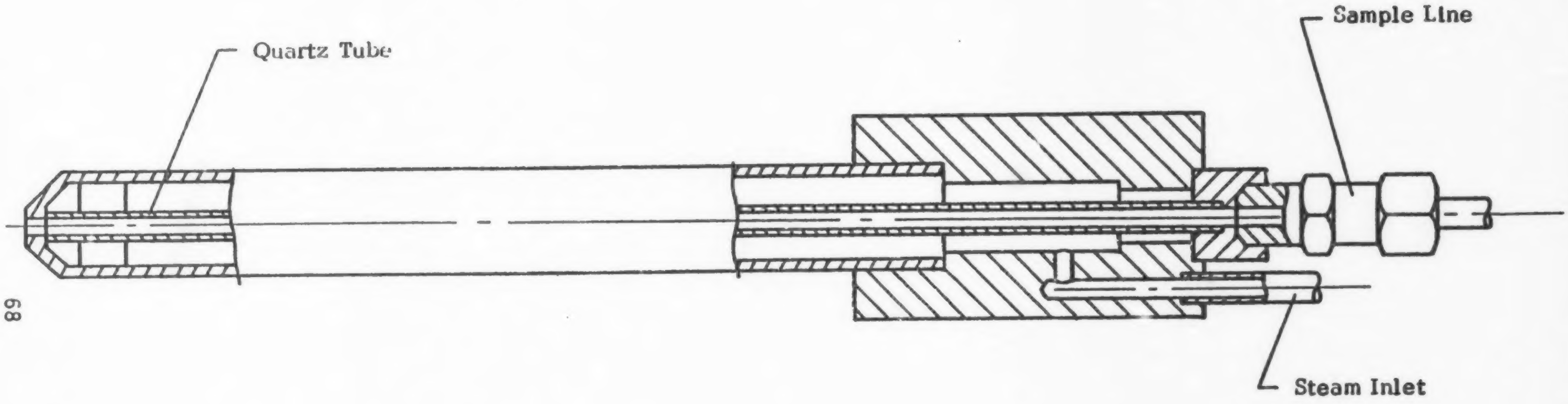


Figure 5.6 A Steam Cooled Quartz Probe For Use In Coal Flames

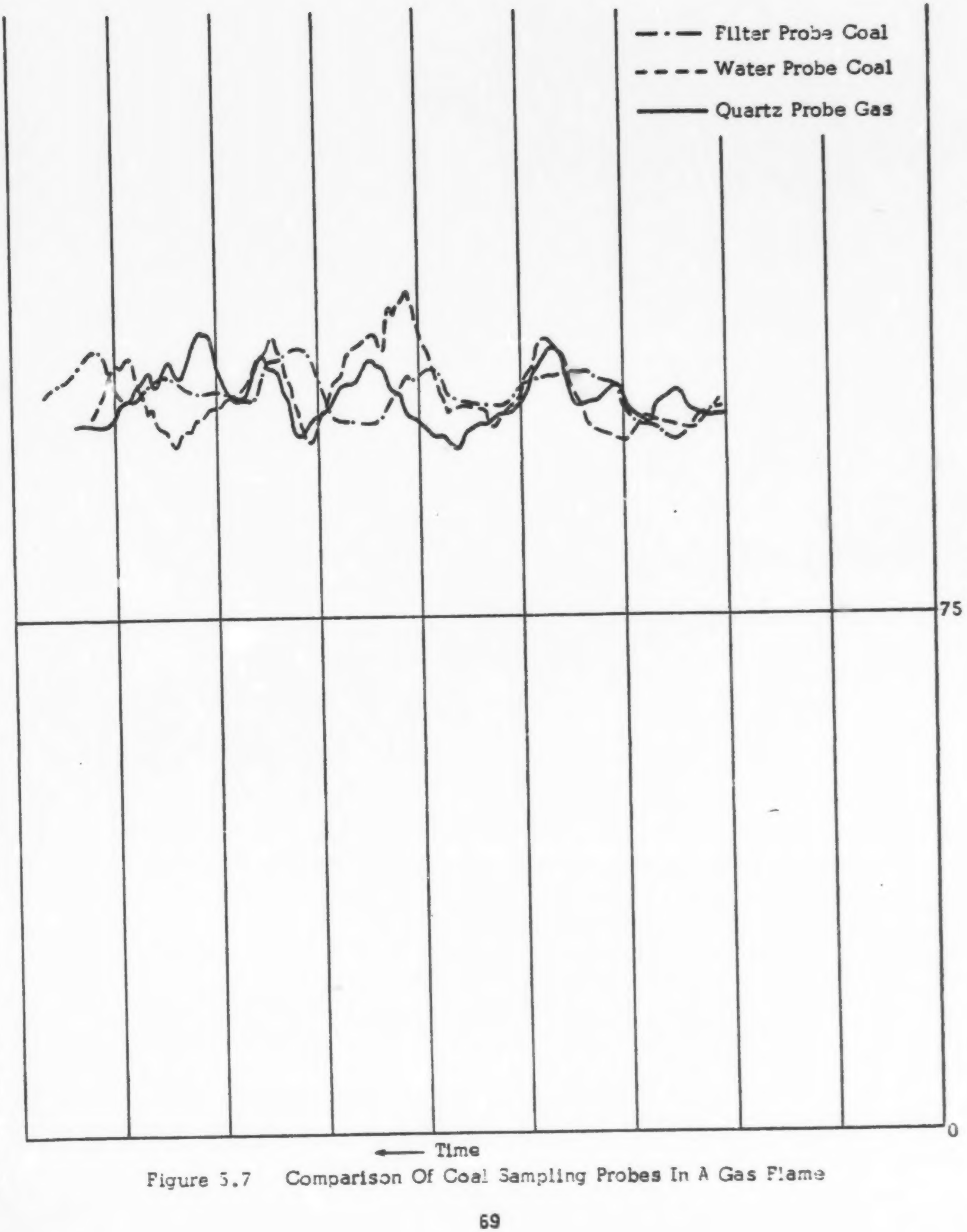


Figure 5.7 Comparison Of Coal Sampling Probes In A Gas Flame

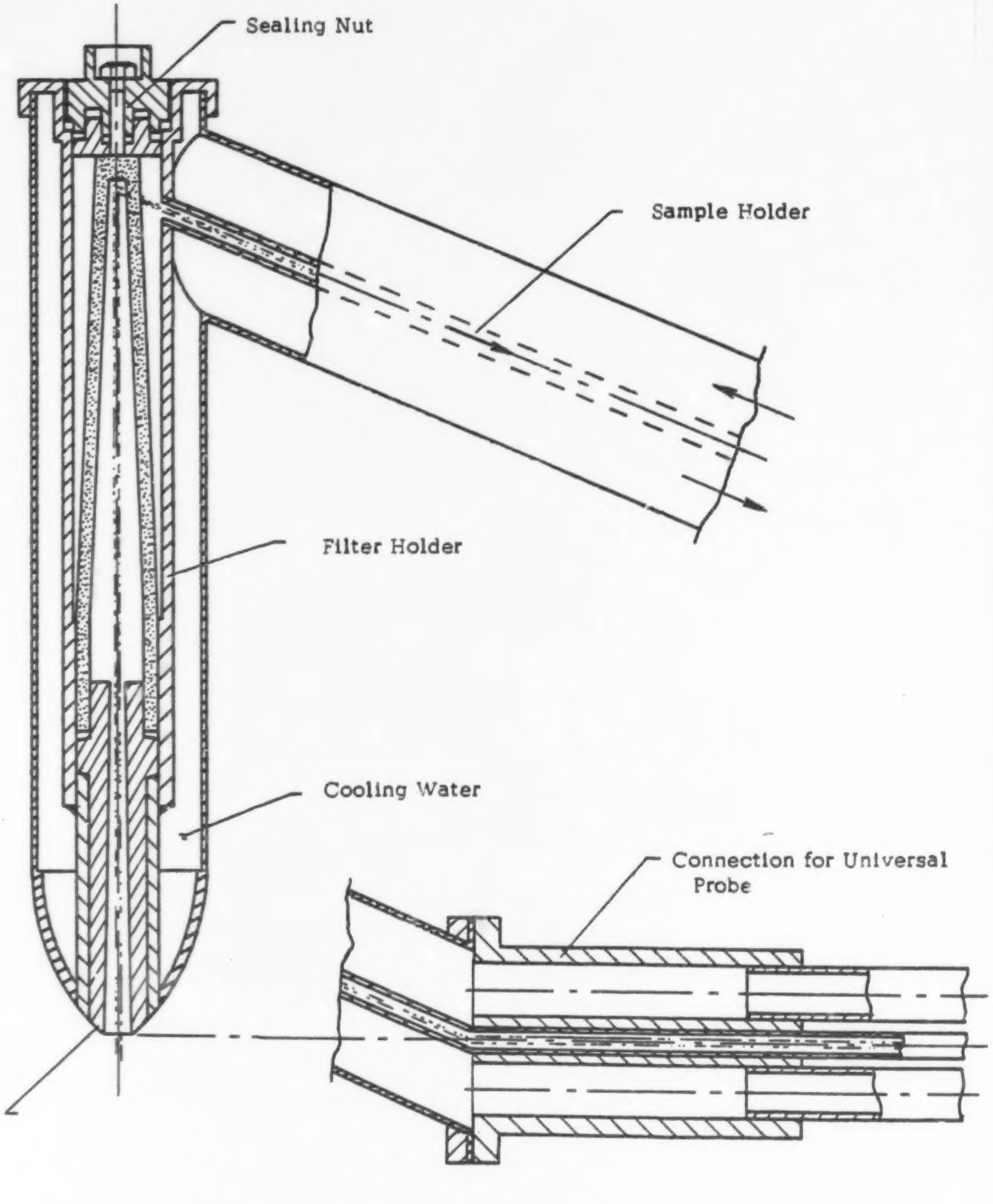


Fig. 5.8 Solid Concentration Sampling Probe (Low Loadings)

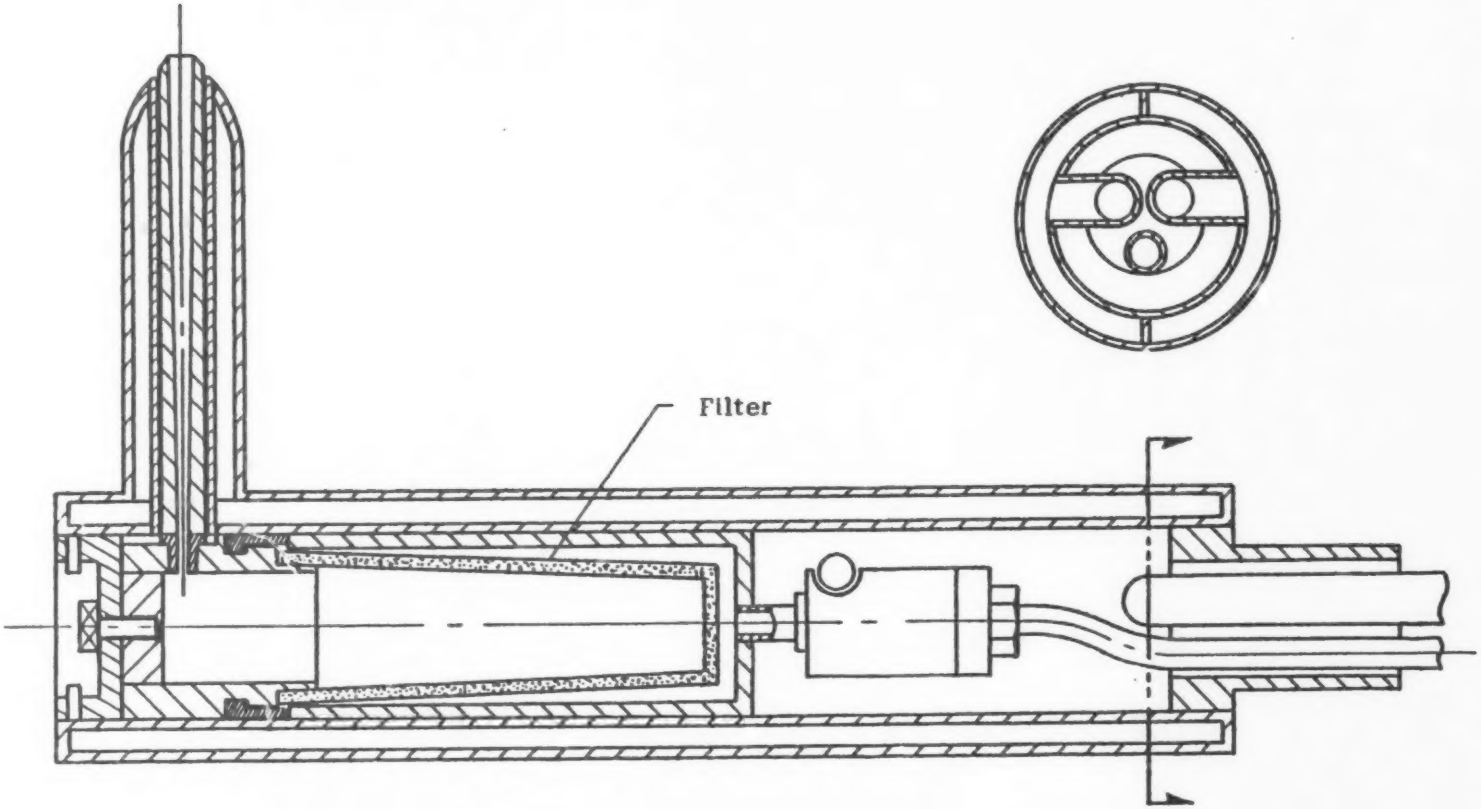


Fig. 5.9 Solid Sampling Probe (High Loadings)

filters were weighed dry before and after use thus enabling the concentration of solids to be calculated if the total volume of gas sampled and the temperature at the sampling point was known. The concentration of the gaseous species were evaluated by gas chromatography from a dry clean sample.

The soot content of the natural gas flames measured during the AP-1 trials was low and was neglected. Consequently, gas samples were taken with a probe without a filter in the sampling head. Thus the water cooled sampling head has considerably reduced dimensions (see Fig. 5.10).

The major problem associated with the sampling of solid matter is that the sample should have the same particle concentration and size distribution as present in the flame. In completely axial flow isokinetic sampling (suction velocity is equal to the gas velocity at the sample point) techniques ensure that these conditions are satisfied. However, in strongly swirling flows a "true sample" is virtually impossible to obtain due to the inertial forces on the particles. Consequently the solid concentrations reported in the appendix cannot be considered as truly representative of the flame conditions.

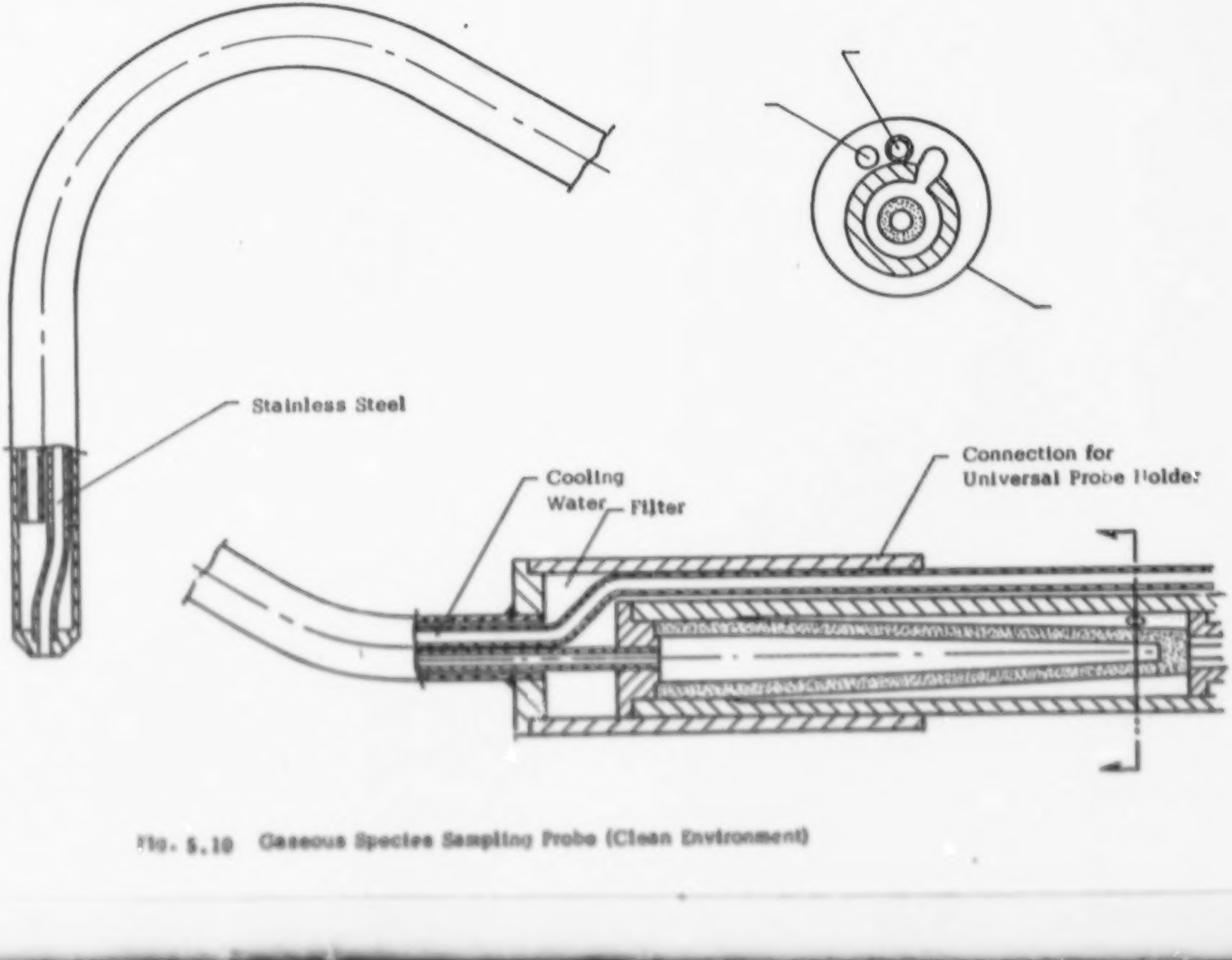


Fig. 5.10 Gaseous Species Sampling Probe (Clean Environment)

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6. RESULTS OF FURNACE INVESTIGATIONS

A complete tabulation of the experimental results is presented in Reference 6.1. In the following sections it is intended that the results of the furnace investigations will be summarized prior to discussing the influence of burner parameters on nitric oxide formation in pulverized coal and natural gas flame.

The major variable involved in the parametric investigations was the degree of rotational swirl in the annular air stream. Throughout the report two methods are used to characterize the degree of swirl:

R_s - relative swirl which is 0 for axial flow and 1.0 at maximum swirl. The intervals correspond to degrees of swirl block adjustment from open to closed; and

S - the dimensionless swirl number (see Section 3).

The relationship between R_s and S can be seen in Fig. 6.1. Relative swirl number is used because in certain instances it allows a comparison to be made more easily between several variables.

6.1 Parametric Investigations Involving Natural Gas

Table 6.1 lists the burner conditions that were investigated with natural gas at a fixed thermal input, excess air rate and preheat level. Fig. 6.2 and Fig. 6.3 give an indication of the range of emission characteristics obtained by varying the fuel injector type, injector position and swirl level. The experimental observations can be summarized as follows:

- increasing the swirl intensity of the combustion air decreased nitric oxide formation in flames produced with multiholed injectors.
- maximum emissions were obtained with the radial hole injector when it was placed at the exit of the burner divergent.
- in almost all instances an increase in throat velocity caused a decreased emission.

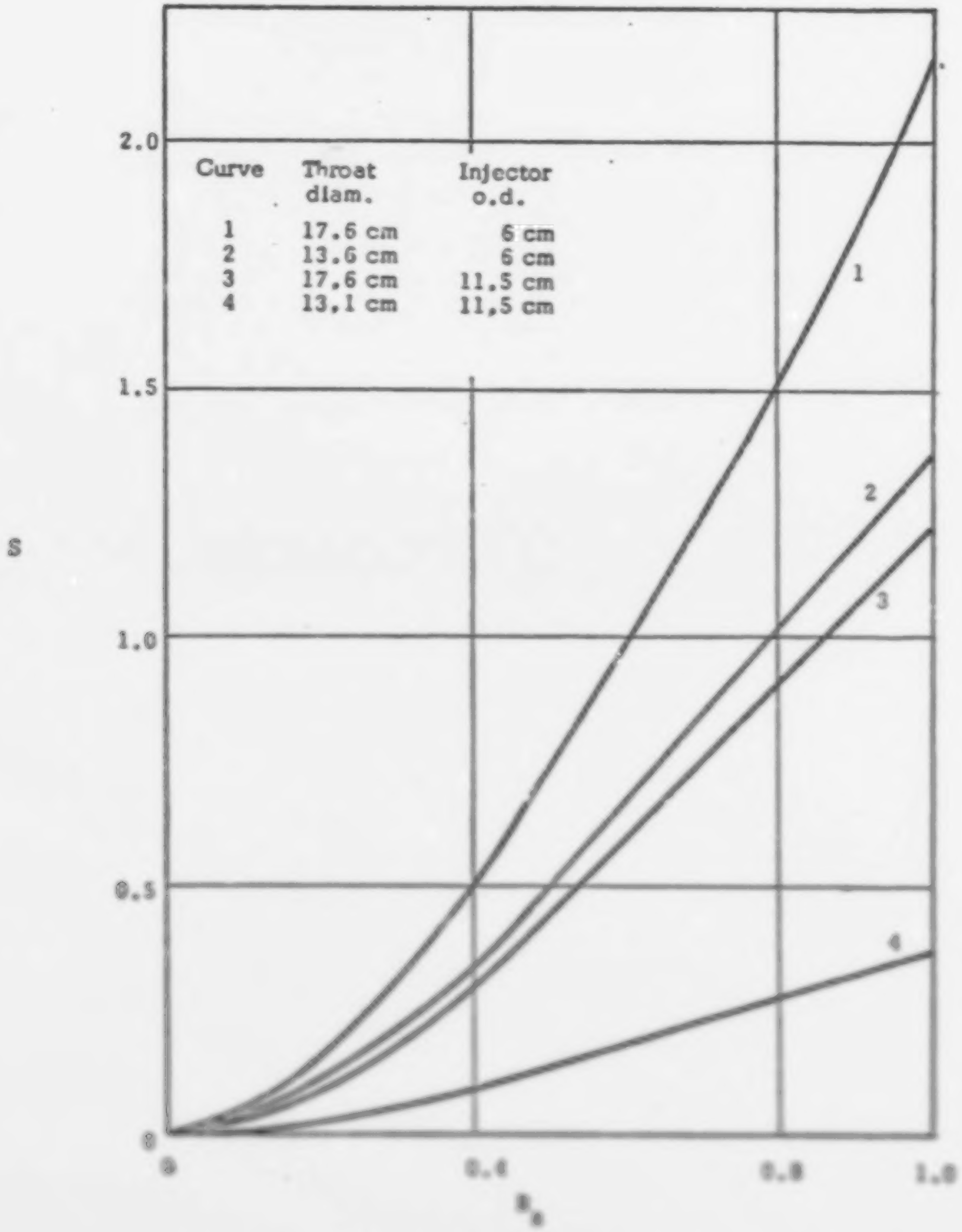

Figure 6.1 The Relationship Between Relative Swirl Index S_a And Swirl Number S

TABLE 6.1 BURNER PARAMETERS INVESTIGATED WITH NATURAL GAS AIR FLAMES (No preheat, 5% excess air 166.13 kgs. hr^{-1} fuel input)

Flame no.	Semi divergent quarl angle	Combustion air velocity	Quarl type	Fuel injector type	Position of injector
1 6 8 10 11	25°	25 msec^{-1}	water cooled	radial hole single hole (1) single hole (2) swirling gas divergent	exit
2 5 7 9 12	25°	25 msec^{-1}	water cooled	radial hole single hole (1) single hole (2) swirling gas divergent	throat
19 21 22 24	25°	50 msec^{-1}	water cooled	divergent single hole (1) single hole (2) radial	throat
20 23	25°	50 msec^{-1}	water cooled	divergent radial	exit
25 26 27 28	35°	50 msec^{-1}	water cooled	radial radial single hole (1) divergent	exit throat
30 31 32	15°	50 msec^{-1}	water cooled	divergent single hole (1) radial	throat
33 34 35	15°	50 msec^{-1}	refractory	radial divergent single hole (1)	throat
36 37 38	35°	25 msec^{-1}	water cooled	single hole (1) radial divergent	throat

• single hole (1) 383 msec^{-1}
single hole (2) 35 msec^{-1}

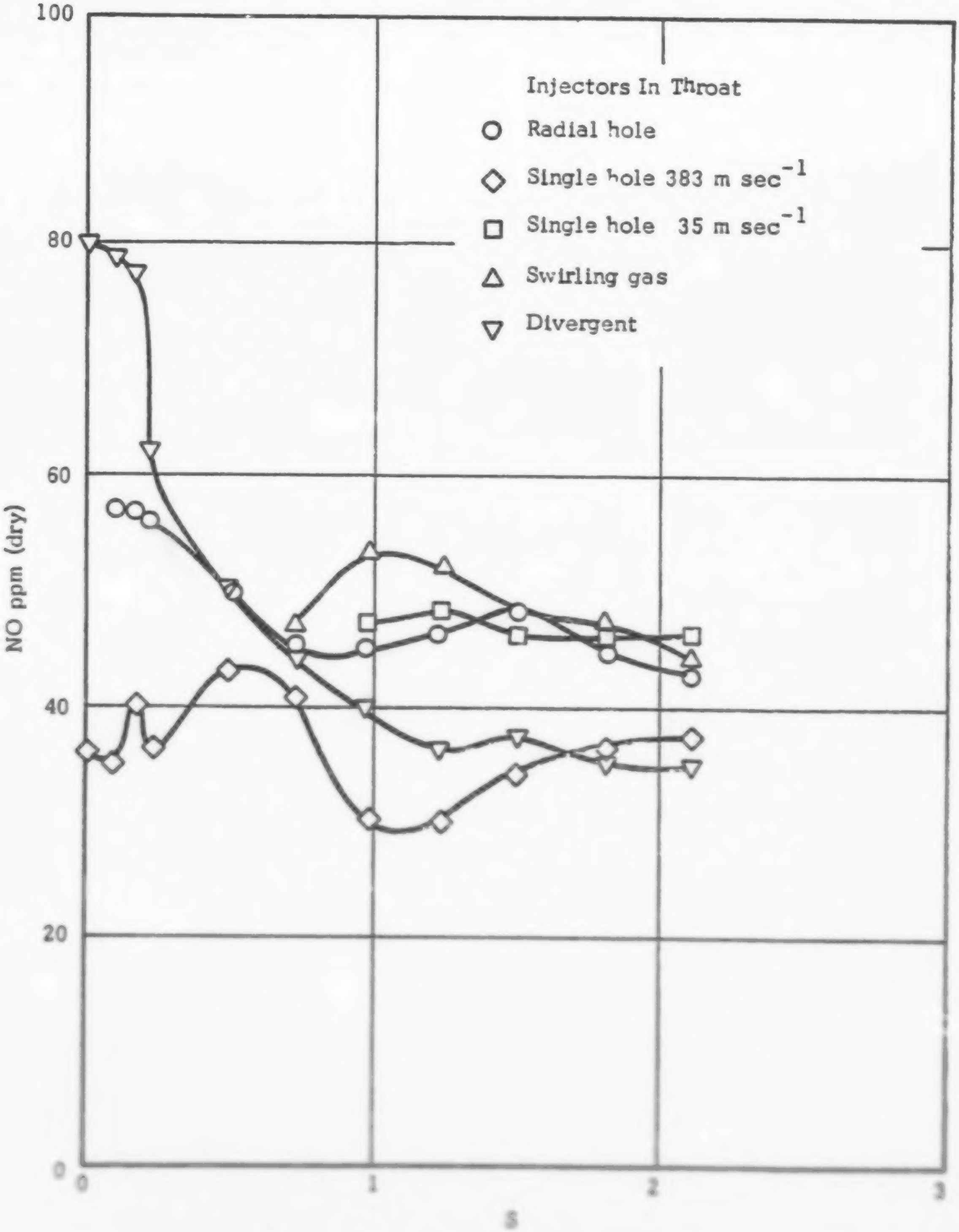

Figure 4.2 Effect Of Fuel Injector Type On NO Emission; Fuel Injector In The Throat Of A 21° Divergent

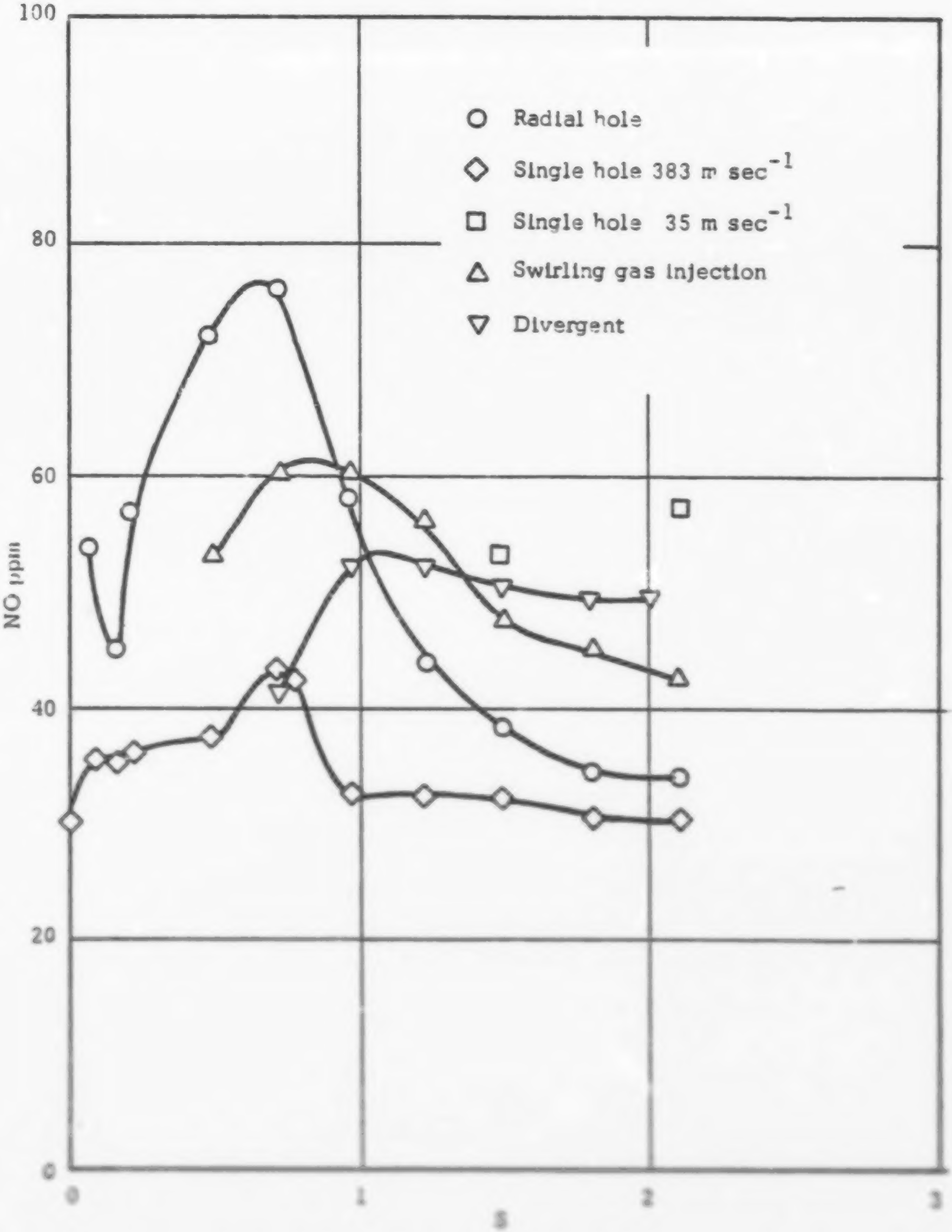

Figure 6.1 Effect Of Fuel Injector Type On NO Emissions: Fuel Injector At The Exit Of A 25° Divergent

- flames stabilized in refractory burner divergents produced more NO than those stabilized in water cooled divergents.

As can be seen from Fig. 6.4 , it appears that burner parameters will also have a strong influence on emission at high elevated preheat levels. Figure 6.5 shows that the general shape of the emission characteristics of radial fuel injectors are unaffected by the excess air level. However, as can be seen from the results presented in Fig. 6.6 the emission level versus excess air peaks at different excess air levels with different swirl intensities. It must be remembered that the increase in excess air will also increase the burner throat velocity.

6.2 Parametric Investigations Involving Pulverized Fuel

Two additional variables were employed in the input/output pulverized coal measurements, the primary air supply and the coal type. The conditions investigated are presented in Table 6.2 a/b. The influence of fuel injector type and swirl can be seen in Fig. 6.7. An attempt will be made to explain the reason for some of the emission curves later in this report. Maximum emissions were observed with radial injectors and minimum emission levels were obtained with high velocity single hole injectors. One important burner variable which was found to influence emissions from pulverized fuel flames was the primary air supply. In some instances the amount of primary air changed the absolute level of the emission curve but not the form of the curve (see Fig. 6.8). However, this was not generally applicable (see Fig. 6.9). Under most burner conditions minimum emissions were observed with 20% primary air supply.

6.3 Detailed Flame Measurements

Six flames were measured in detail during the furnace investigations and their input conditions are listed in Table 6.3. Complete measurements details are presented in Reference 6.1.

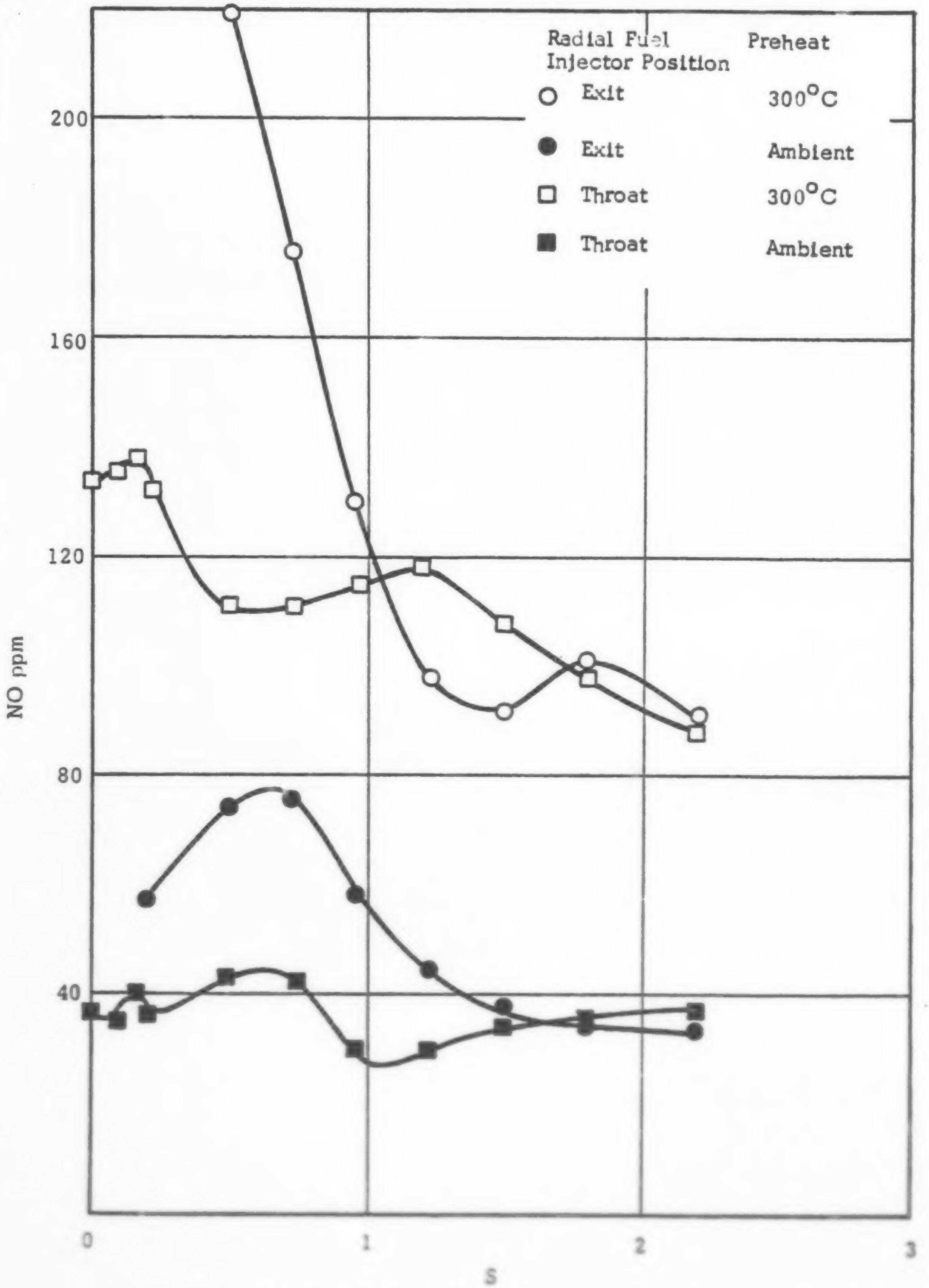

Figure 6.4 The Effect Of Preheat On The Emission Of Nitric Oxide (Radial Fuel Injector, 5% excess Air)

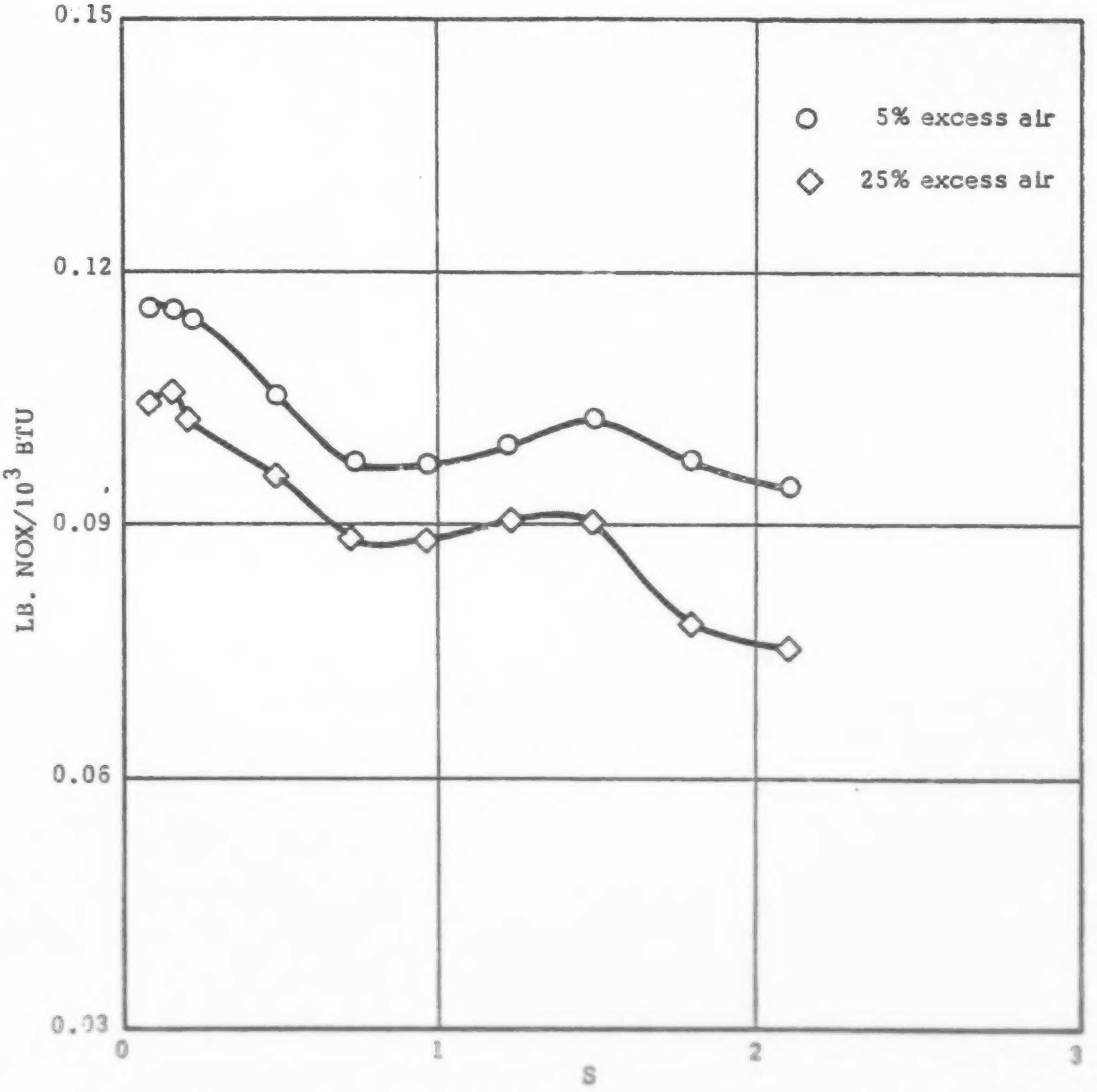

Figure 6.5 The Influence Of Excess Air On NO Emission Radial Injector In The Throat Of A 25° Divergent

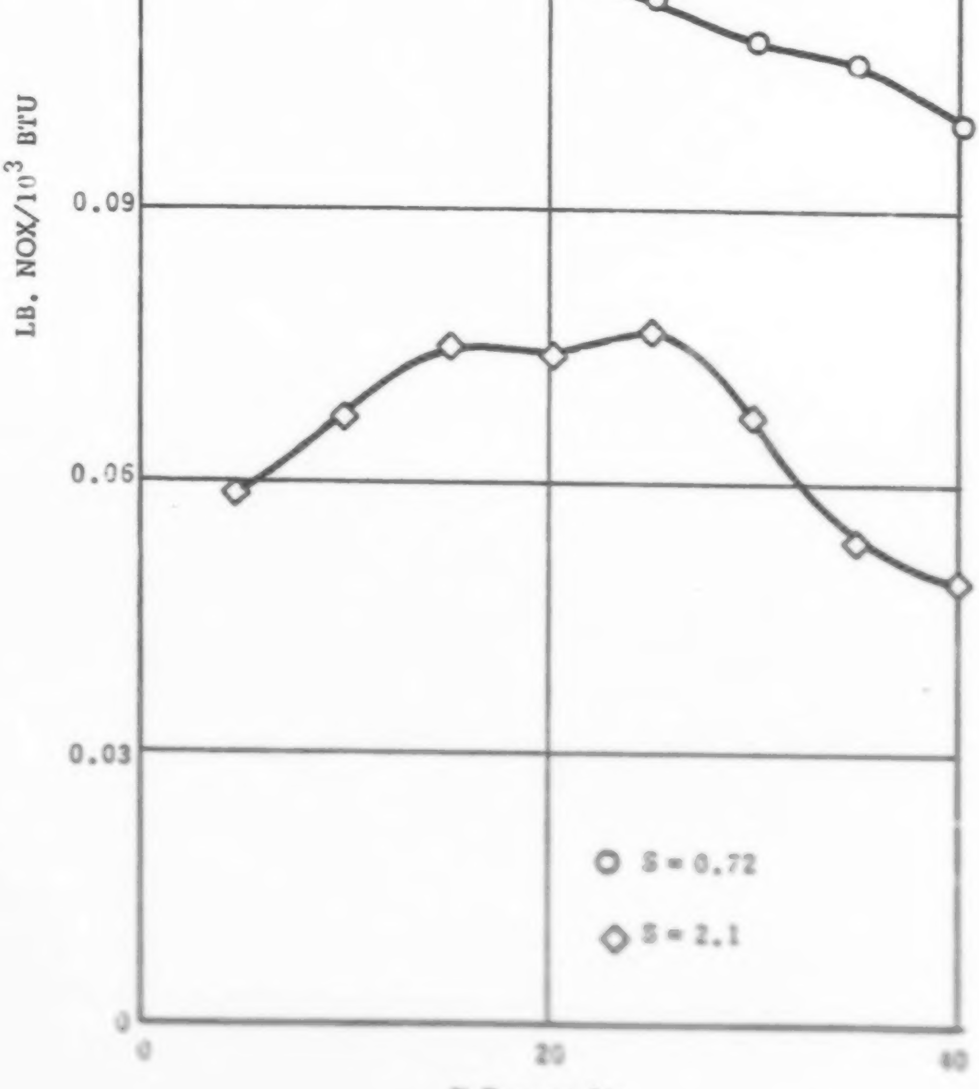

Figure 6.6 The Effect Of Excess Air On NO Emission At Two Swirl Levels. Radial Injector At The Exit Of A 25° Divergent

TABLE 6.2a CONDITIONS INVESTIGATED DURING THE PULVERIZED COAL INPUT/ OUTPUT SECTION OF THE AP-1 TRIALS

Flame no.	burner	excess air	throat	position	P_{prim}	Quarl L	Coal $\%N_2$
86	A	5%	176	throat	10%	0	1.1%
63	"	"	"	"	"	25	"
85	"	"	"	exit	"	0	"
70	"	"	"	"	"	25	"
103	"	"	131	"	"	0	"
88	"	"	176	throat	20%	0	"
66	"	"	"	"	"	25	"
43	"	"	"	"	"	35	"
83	"	"	"	exit	"	0	"
69	"	"	"	"	"	25	"
102	"	"	131	"	"	0	"
104	"	"	"	"	"	25	"
87	"	"	176	throat	30%	0	"
67	"	"	"	"	"	25	"
44	"	"	"	"	"	35	"
84	"	"	"	exit	"	0	"
68	"	"	"	"	"	25	"
106	"	"	131	throat	"	25	"
101	"	"	131	exit	"	0	"
105	"	"	"	"	"	25	"
73	B	"	176	throat	10%	25	"
95	"	"	"	exit	"	0	"
74	"	"	"	"	"	25	"
98	"	"	131	"	"	0	"
71	"	"	176	throat	20%	25	"
96	"	"	"	exit	"	0	"
75	"	"	"	"	"	25	"
108	"	"	131	throat	"	0	"
109	"	"	"	"	"	25	"
99	"	"	"	exit	"	0	"
72	"	"	176	throat	30%	25	"
97	"	"	"	exit	"	0	"
76	"	"	"	"	"	25	"
100	"	"	131	"	"	0	"

TABLE 6.2a (Continued)

Flame no.	burner	excess air	throat	position	P_{prim}	Quarl, L	Coal %N_2
53	C	5%	176	throat	10%	25	1.1
45	"	"	"	"	"	35	"
94	"	"	"	exit	"	0	"
57	"	"	"	"	"	25	"
46	"	"	"	throat	20%	35	"
93	"	"	"	exit	"	0	"
47	"	"	"	throat	30%	35	"
79	D	"	"	throat	20%	25	"
80	"	"	"	"	30%	"	"
91	E	"	"	exit	10%	0	"
89	"	"	"	"	20%	"	"
90	"	"	"	"	30%	"	"
78	F	"	"	throat	10%	25	"
77	"	"	"	"	20%	"	"
82	G	"	"	throat	20%	25	"
81	"	"	"	"	30%	"	"
60	H	"	"	throat	10%	25	"
92	"	"	"	exit	"	0	"
59	"	"	"	"	"	25	"
138	A	"	"	throat	10%	25	1.8
134	"	"	"	exit	"	"	"
142	"	"	"	throat	20%	"	"
126	"	"	"	exit	"	"	"
146	"	"	"	throat	30%	"	"
130	"	"	"	"	"	"	"
150	E	"	"	throat	20%	25	"
151	"	"	"	exit	"	"	"
152	"	"	"	throat	30%	"	"
153	"	"	"	exit	"	"	"

TABLE 6.2a (Continued)

Flame no.	burner	excess air	throat	position	P_{prim}	Quarl L	Coal %N_2
154	D	5%	176	throat	20%	25	1.8
156	"	"	"	"	30%	"	"
160	F	"	"	"	10%	"	"
158	"	"	"	"	20%	"	"
40	A	20%	176	throat	30%	35	1.1
41	"	"	"	"	10%	"	"
42	"	"	"	"	20%	"	"
64	"	"	"	"	10%	25	"
65	"	30%	"	"	10%	25	"
51	F	20%	"	"	20%	35	1.1
52	"	"	"	"	10%	"	"
48	C	20%	"	"	20%	35	"
49	"	"	"	"	10%	35	"
50	"	"	"	"	30%	35	"
54	"	15%	"	"	10%	25	"
55	"	20%	"	"	"	"	"
56	"	30%	"	"	"	"	"
58	"	30%	"	exit	"	"	"
127	A	10%	"	exit	20%	25	1.8
128	"	20%	"	"	"	25	"
129	"	30%	"	"	"	"	"
131	"	10%	"	"	30%	"	"
132	"	20%	"	"	"	"	"
133	"	30%	"	"	"	"	"
135	"	10%	"	"	10%	"	"
136	"	20%	"	"	10%	"	"
137	"	30%	"	"	"	"	"
139	"	10%	"	throat	"	"	"
140	"	20%	"	"	"	"	"
141	"	30%	"	"	"	"	"

TABLE 6.2a (Continued)

Flame no.	burner	excess air	throat	position	P_{prim}	Quarl L	Coal %N
143	A	10%	176	throat	20%	25	1.0
144	"	20%	"	"	"	"	"
145	"	30%	"	"	"	"	"
147	"	10%	"	"	30%	"	"
148	"	20%	"	"	"	"	"
149	"	30%	"	"	"	"	"
155	D	20%	"	"	20%	"	"
157	"	"	"	"	30%	"	"
159	F	"	"	"	20%	"	"

TABLE 6.2b COAL BURNER CHARACTERISTICS

Burner	Type Of Injection	Dimensions	Mean Injection Velocity m/sec:			Secondary* Air Velocity
			10% Primary Air	20% Primary Air	30% Primary Air	
A	Single hole	54.5 mm id 115 mm od	19	38	57	High
B	Single hole	54.4 mm id 60 mm od	19	38	57	Low
C	Single hole	46 mm id 60 mm od	26	52	-	Low
D	Annular	44.5 mm x 107 mm 115 mm od	-	12	18	High
E	Single hole	70 mm id 115 mm od	11	22	33	High
F	Radial	16 holes 60 mm od	27	54	-	Low
G	Single hole	64 mm id 115 mm od	14	28	42	High
H	Single hole	33 mm id	52	-	-	Low

*Actual Velocity Dependent upon primary air flow.

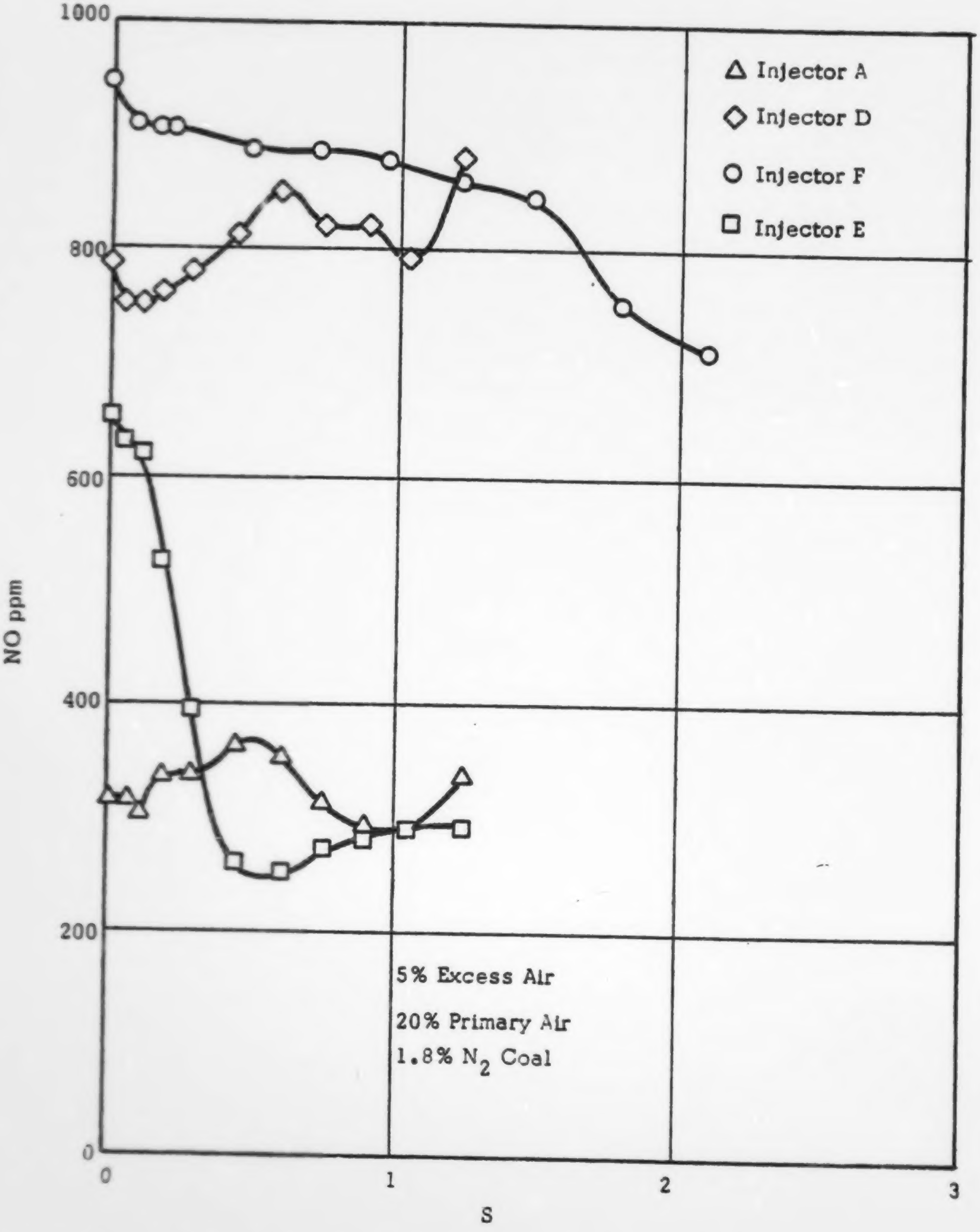

Figure 6.7 The Effect Of Fuel Injector Type And Swirl On NO Formation In P.F. Flames

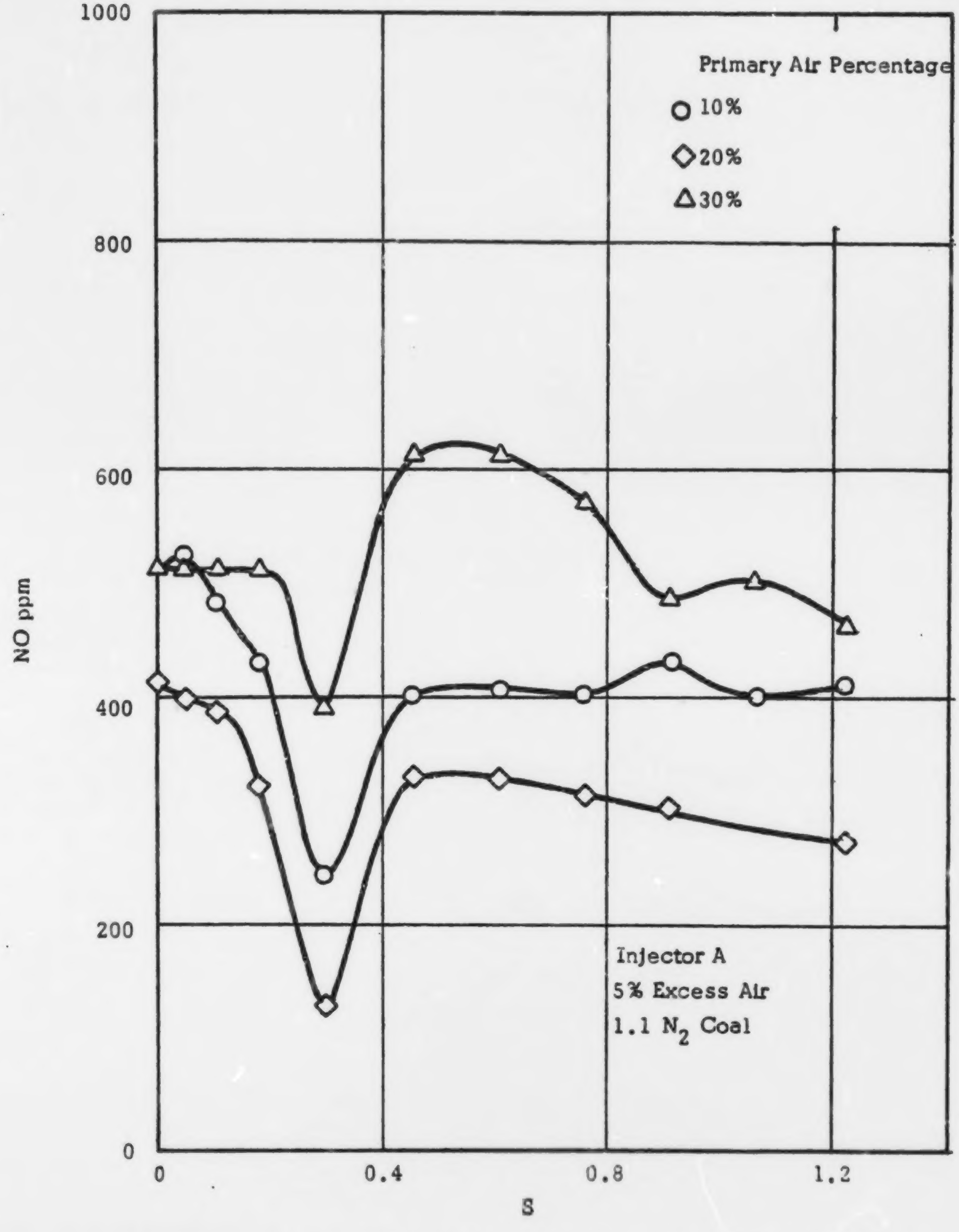

Figure 6.8 The Effect Of Primary Air Percentage On NO Formation In Coal Flames

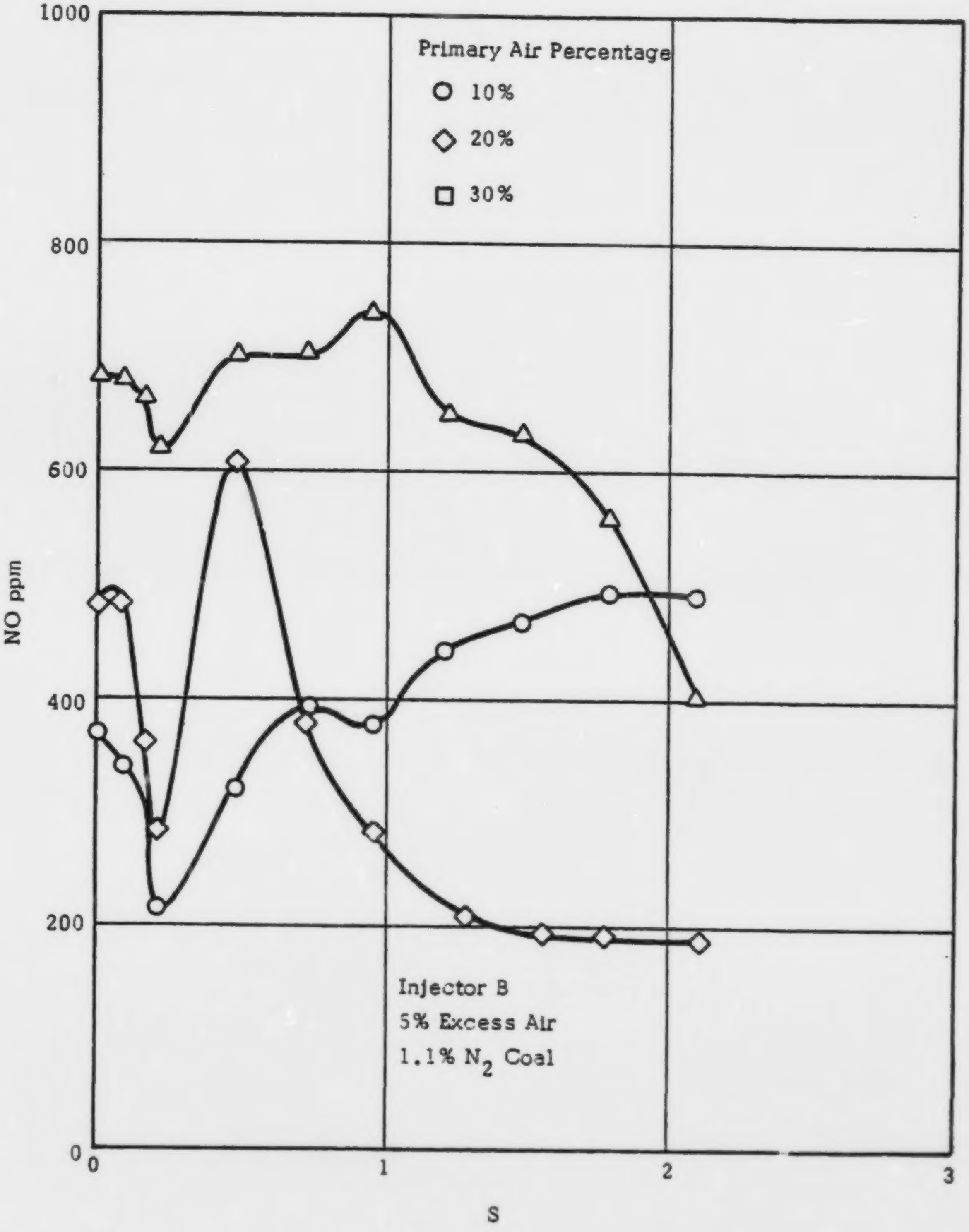

Figure 6.9 The Effect Of Primary Air Percentage On NO Formation In Coal Flames

TABLE 6.3 INPUT CONDITIONS FOR MAIN FLAME MEASUREMENTS

Flame No.	Fuel	Injector Type	Position	R_s	Primary Air	Excess Air
119	gas	33	throat	.6	-	5%
120	gas	8	throat	.9	-	5%
121	coal	F	throat	.6	20%	25%
122	coal	B	throat	.9	20%	25%
123	coal	A	exit	.9	30%	10%
124	coal	A	exit	.9	20%	10%

For all flames burner divergent 25°, and throat diameter 17.6 cms, 300°C air preheat.

REFERENCES

6.1 Heap, M.P., Lowes, T.M., Walmsley, R., Bartelds, H., and Le Vaguerese, P., Appendix to Volume I - Investigations to define the influence of burner variables upon nitric oxide formation in natural gas and pulverized coal flames - Experimental Results (to be published).

7. DISCUSSION OF RESULTS — NATURAL GAS FLAMES

The influence of burner variables on nitric oxide formation in natural gas flames has been studied in a series of parametric investigations which were described in Ssection 6. The range of emission levels measured with the different variables can be seen in Fig. 7.1 when plotted as a function of swirl number. Fig. 7.1 also indicates the range of emission levels for the various characteristics flame types. At present there is no fundamental model which is capable of predicting the effect of burner variables on emission levels. However, some kind of mechanistic understanding must be obtained if the results are to be used to specify design criteria for low emitting burners.

7.1 A Qualitative Model of Nitric Oxide Formation in Turbulent Gaseous Flames

The natural gas used in this investigation does not contain chemically-bound nitrogen, although approximately 10-14 percent of molecular nitrogen is present in the gas. Consequently, only the formation of thermal NO need be considered.

The discussions presented in Section 3 emphasized the strong influence of temperature on the rate of formation of NO. Figure 7.2 presents axial nitric oxide concentrations for four flames. The corresponding axial temperature distributions are presented in Fig. 3.5 which also shows the maximum measured temperatures. It can be seen that for none of the flames does the measured temperature exceed 1550°C. Also, the maximum temperatures measured for the two jet flames are very similar and yet the emission of the flame with weak swirl is about half that of the flame with no swirl. When no preheat is used external recirculation temperatures do not exceed 1200°C. Thus, for the flames under discussion in this report, nitric oxide formation can be considered solely as a flame phenomenon since insignificant amounts of nitric oxide will be formed in the bulk gases. The nitric oxide distributions shown in Fig. 7.2 cannot be accounted for on the basis of the flame temperatures shown in Fig. 3.5. These temperatures are time mean values. Neither the response time of the suction pyrometer nor the spatial resolution are sufficient to measure the temperatures which are believed to be of importance for NO formation in turbulent diffusion flames (7.1).

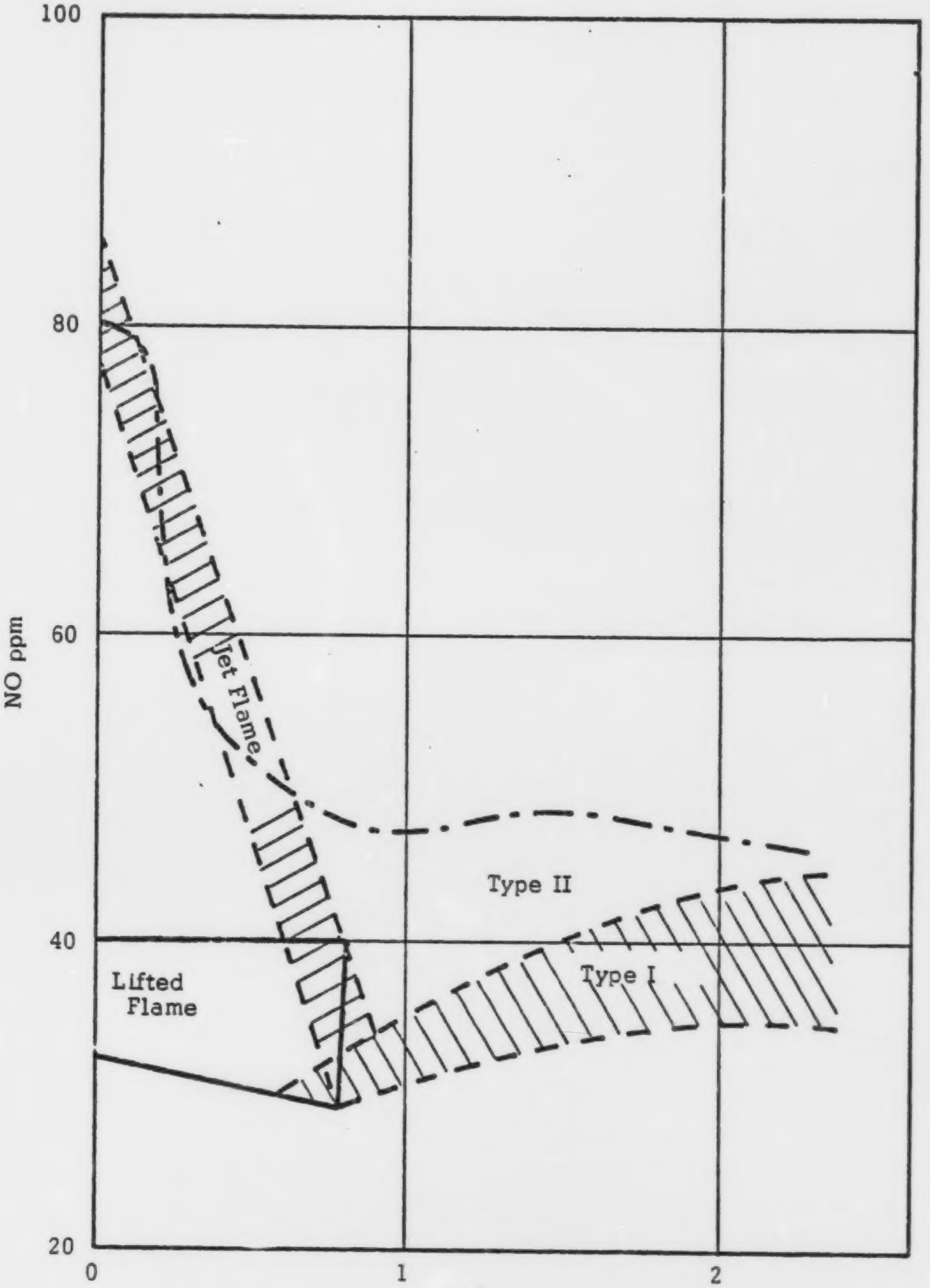

Figure 7.1 NO Emission Characteristics Of The Various Flame Types

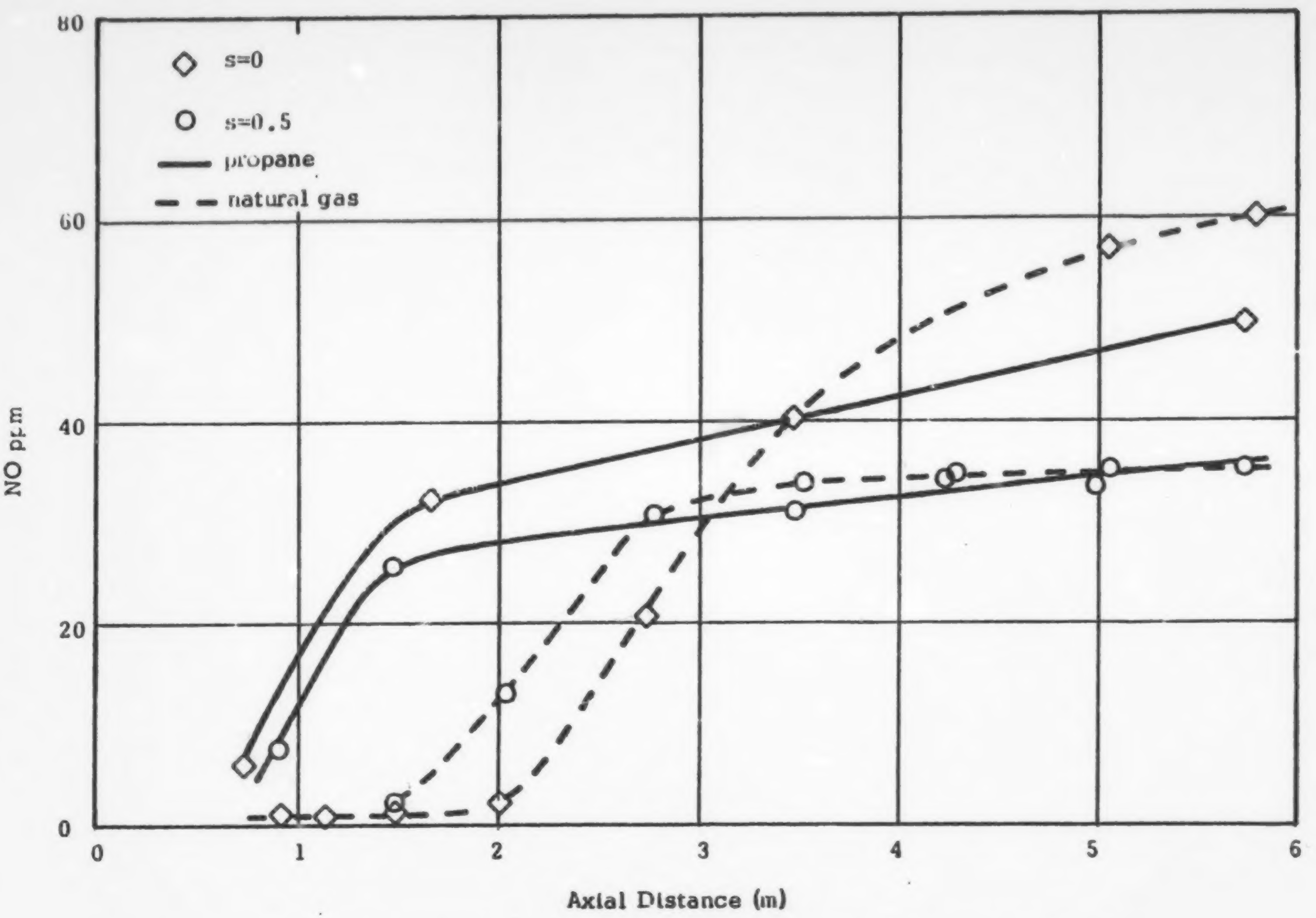

Figure 7.2 The Influence Of Swirl An Axial NO Distribution Of Jet Flames

In turbulent diffusion flames the total heat release is distributed over discrete zones. The sum of the zone volumes is less than the "time mean" total flame volume which consists of:

- unmixed fuel air and combustion products;
- mixed but unreacted fuel air and combustion products;
- reacting fuel and air;
- products of combustion.

The amount of nitric oxide produced from the combustion products associated with one element of fuel will depend upon:

- their initial temperature T_B;
- the rate of change of temperature from T_F to some cutoff temperature T^1 below which the rate of NO production becomes insignificant;
- the true oxygen atom concentration.

Although kinetic studies have indicated the importance of the last factor it will be neglected in further discussion and it will be assumed that the temperature T_F and the function $\frac{dT}{dt}$ control NO production in turbulent diffusion flames.

The total heat released within the flame volume is fixed at the input rate Q which is the sum of all the heat release rates within the discrete reaction zones, this

$$Q = \sum_{x=1}^{x=r} q_x \tag{21}$$

where q_x represents the heat release rate of one reaction zone. Obviously the number of the individual zones will vary with time depending upon the turbulent mass transfer process. Two essentially different types of reaction zones can be envisaged:

- mixing between fuel and air takes place before the reactant temperature is sufficient to ensure that reaction will take place. Thus, provided the mixture composition lies within the limits of flamability, if reaction is initiation at any one point a flame can propagate throughout the entire mixture;

- due to mixing with combustion products either of the reactants are at a sufficient temperature to allow reaction to proceed should mixing occur.

The temperature T_F of the freshly formed products of combustion will depend on the composition and enthalpy of the mixture prior to reaction. It is important to note that T_F does not depend on the fuel air equivalence ratic alone, but also on the degree of dilution by and the temperature of any combustion products. Maximum values of T_F will occur when the fuel and air are undiluted with combustion products react in proportions near to stoichiometric. The rate at which the temperature of the freshly formed products of combustion will be reduced depends upon:

- the bulk gas temperature;
- the rate of mixing with the bulk gases;
- the amount of radiative heat loss which will be dependent on the volume and emissivity of the combustion products and the temperature of the furnace enclosure and the bulk gases.

Heat loss by radiation will play only a minor role. The major influence on the rate of temperature decrease will be the mixing rate between the bulk gases and the flame gases. Should the reacting gases come close to cooled surfaces then convective heat transfer could also be important. Burner variables influence the amount of nitric oxide produced in natural gas flames because they control the mixing history of the four components of interest:

- the fuel;
- the air;
- internally recirculating combustion products;
- externally recirculating combustion products;

The mixing history of these components will dictate the degree of dilution and the reaction zone equivalence ratio and therefore the temperature T_F. Also the rate of mixing will influence the rate of temperature decrease of the products of combustions.

The influence of burner variables on the mixing history of the four separate components can be summarized by:

- the type of fuel injector and its position will affect both the fuel air mixing pattern and the mass exchange between the fuel and internally recirculation vortex;

- the degree of swirl will control the rate of entrainment of external recirculation and the size, intensity and position of the internal recirculation vortex;
- the throat velocity will influence the fuel air mixing pattern and the rate of entrainment of external recirculation;
- the geometry of the burner exit will influence the size and intensity of the recirculating gases.

With a knowledge of the properties of swirling flames it is possible to use the qualitative model describing the formation of nitric oxide to explain the influence of burner variables on the emission characteristics of large scale natural gas flames.

7.2 Burner Variables and Nitric Oxide Formation

7.2.1 Axial Fuel Jet Flames

Axial fuel injectors of sufficient velocity can produce:

- lifted flames;
- simple jet flames with $0 < S <$ critical;
- high intensity type I flames.

Due to stability problems the characteristics of simple jet flames could only be investigated with oxygen enriched air. In general it was found that weak swirl reduced emissions (See Fig. 7.2). Radial distributions of temperature and nitric oxide are presented in Fig. 7.3 and Fig. 7.4 for the two jet flames. As expected the nitric oxide measured on the axis is a minimum. The NO measured in the forward jet is the sum of that produced in the flame and that entrained with the external recirculation. Both the axial and radial profiles presented for the two flames emphasize the fact that the measured time mean temperatures are too low to produce NO. Also the measured profiles show temperatures of similar magnitude even though the final emission level was reduced from 60 to 35 ppm when the swirl number was increased from 0 to 0.5.

The reduced emission from simple jet flames with the application of swirl can be attributed to two factors associated with improved mixing

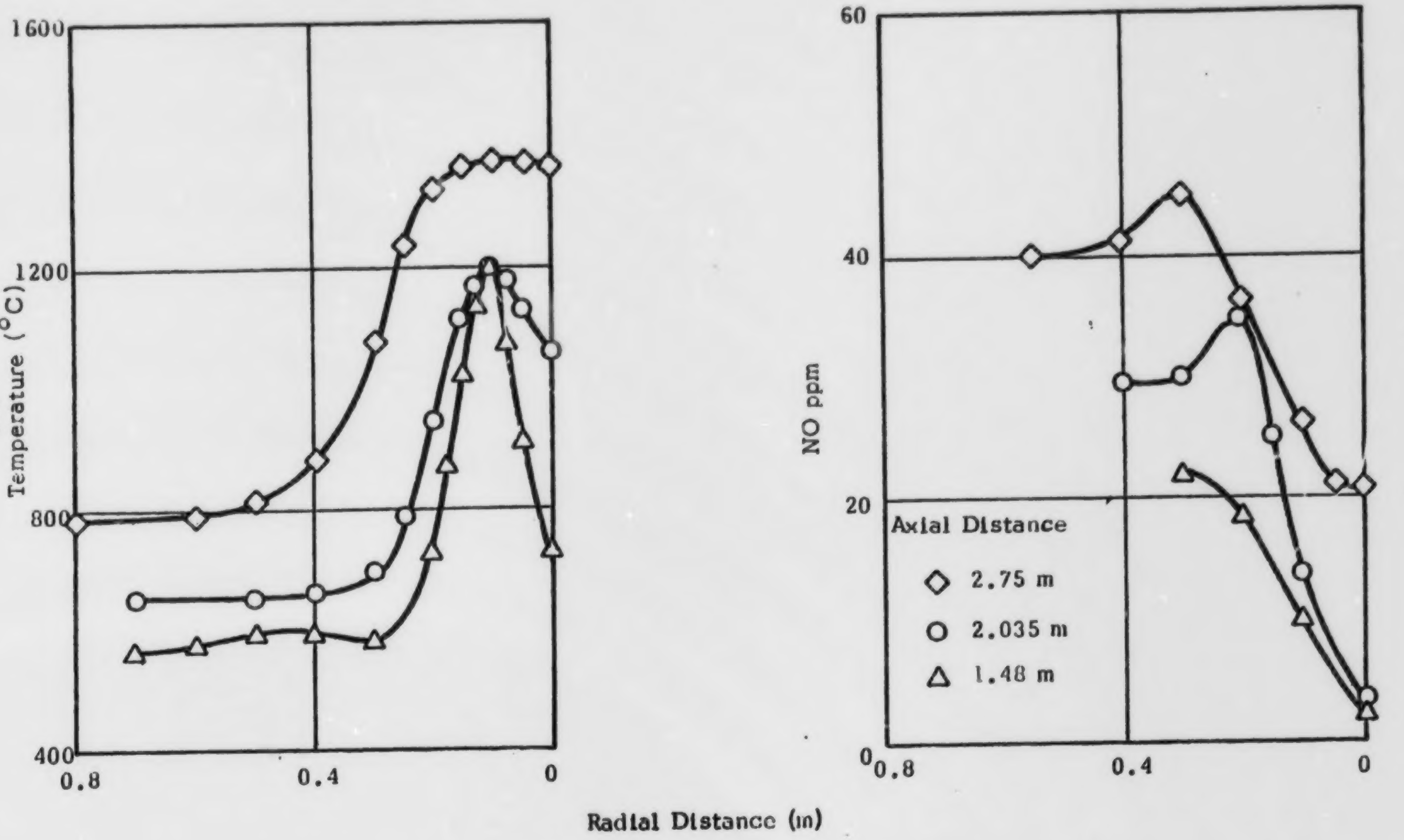

Figure 7.3 Radial Temperature And NO Distribution For Non-Swirling Jet Flames

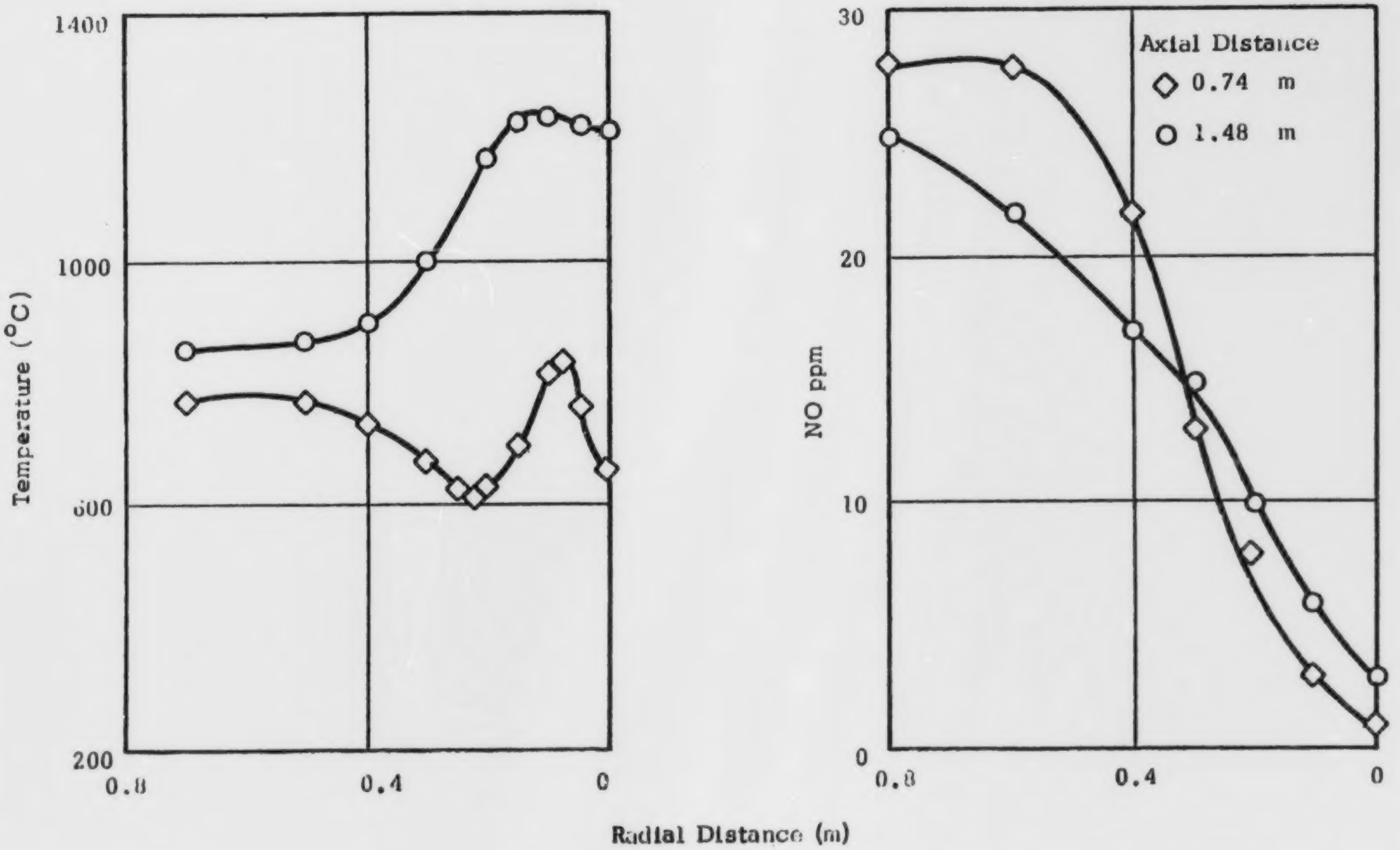

Figure 7.4 Radial Temperature And NO Distributions For Slightly Swirling (s=0.5) Jet Flame

- increased entrainment of external recirculation increases the rate of decrease of temperature from T_F to T^1 of the combustion products;

- increased entrainment will mean that the dilution of the reactants prior to combustion will increase thus reducing T_F.

From visual observations it is known that the application of swirl spreads the flame and the flame length is decreased. The increased rate of entrainment of the swirling jet can be seen from the results presented in Fig. 7.5 which compares the forward mass flow for the two flames. Similar measurements have been made in two propane flames. Swirl also reduces the emission of nitric oxide from propane jet flames although the reduction is not so startling. Fig. 7.6 shows both the axial temperature and NO distributions for the two flames. As with natural gas the applications of swirl spreads the flame and the axial temperature gradient is steeper although the measured temperatures with and without swirl are similar. The most significant difference, associated with nitric oxide formation with different fuel gases is their radial NO distributions. Compare the radial profiles presented in Figs. 7.7 and 7.8 for propane with those in Figs. 7.3 and 7.4. Both propane curves have a maximum on the axis while with natural gas the curves have a minimum on the axis. Although the temperature profiles were similar for comparable flames with different fuels species concentrations were different (See Fig. 7.9). Larger concentrations of hydrogen, carbon monoxide and less oxygen were measured on the axis of propane flames than on natural gas flames. Visually the propane flames were more luminous than their natural gas counterparts.

The observations described above can be attributed to:

- measurement errors. Soot deposited at the tip of the water-cooled quartz sampling probe may react with the sampled gases producing erroneous results;

- the nitric oxide formed within the fuel rich core of the propane flame may be produced by the "prompt NO" mechanism suggested by Fenimore (7.1).

The above arguments are speculative and more detailed measurements should be carried out to ascertain whether or not the effects observed with propane jet

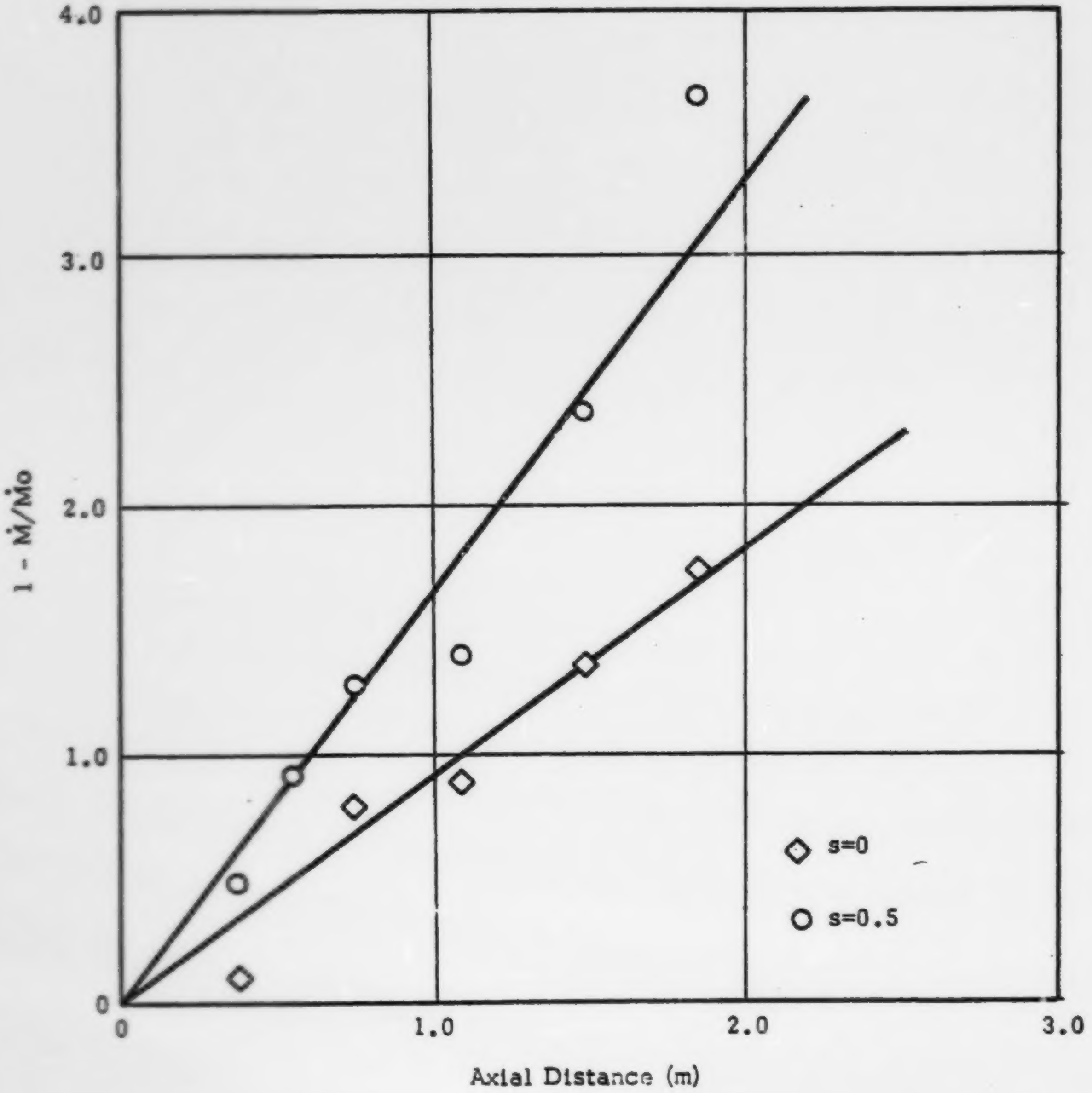

Figure 7.5 Comparison The Forward Mass Flow In The Early Regions Of Non-Swirling And Slightly Swirling Flames

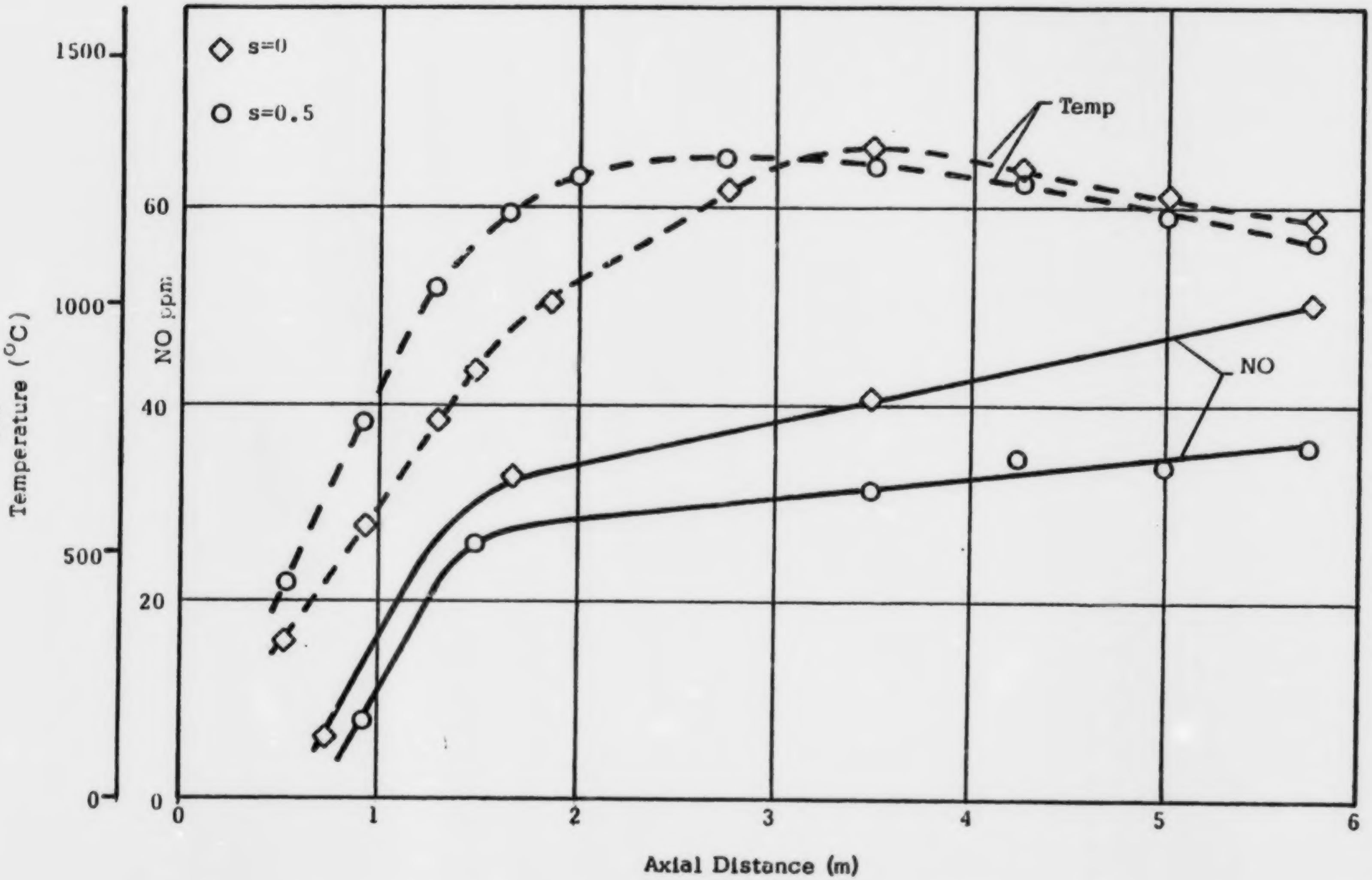

Figure 7.6 Axial Temperature And NO Distribution For 3MW Propane Oxygen Nitrogen Flames

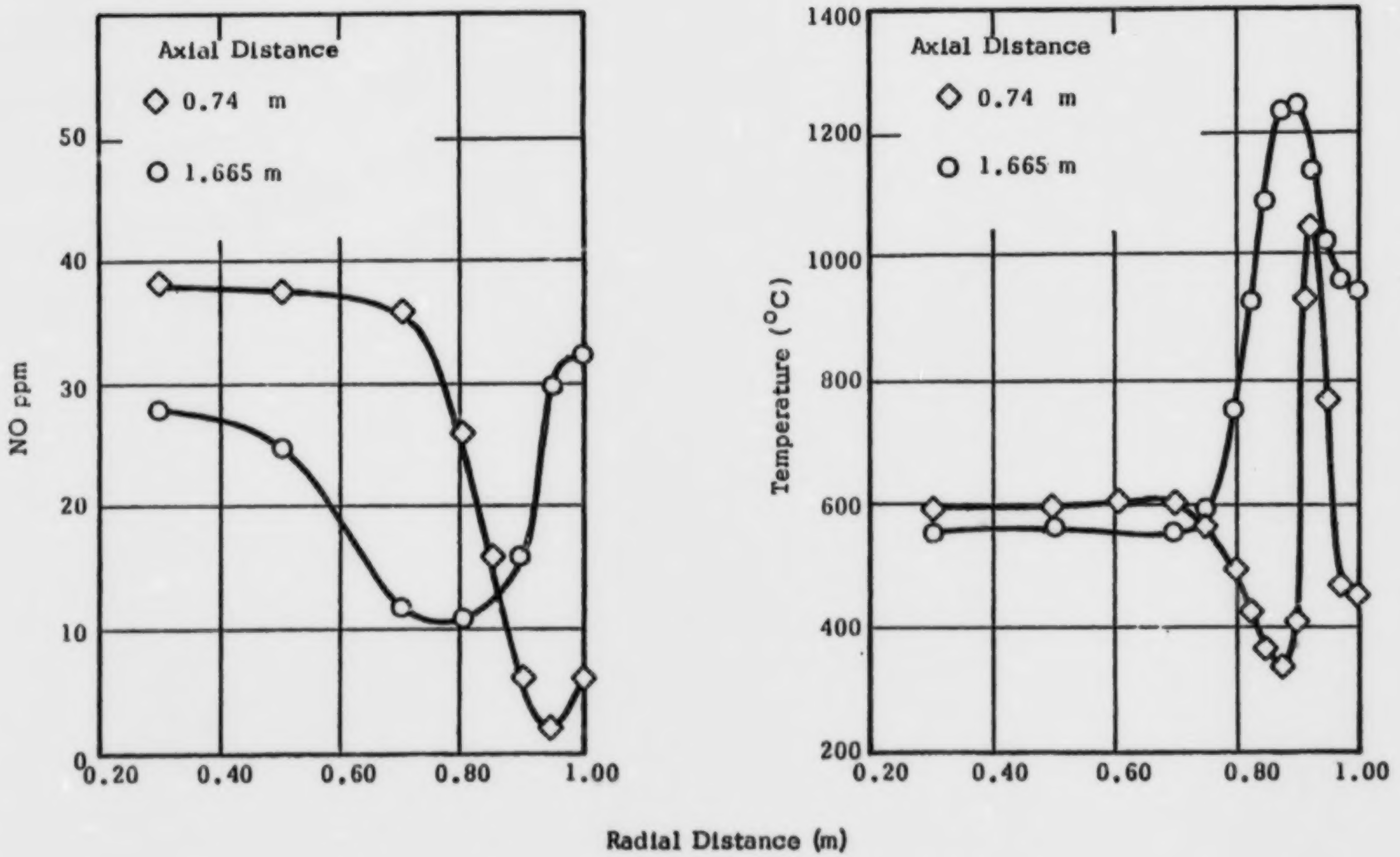

Figure 7.7 Radial Temperatue And NO Distributions For Non Swirling Propane Flame

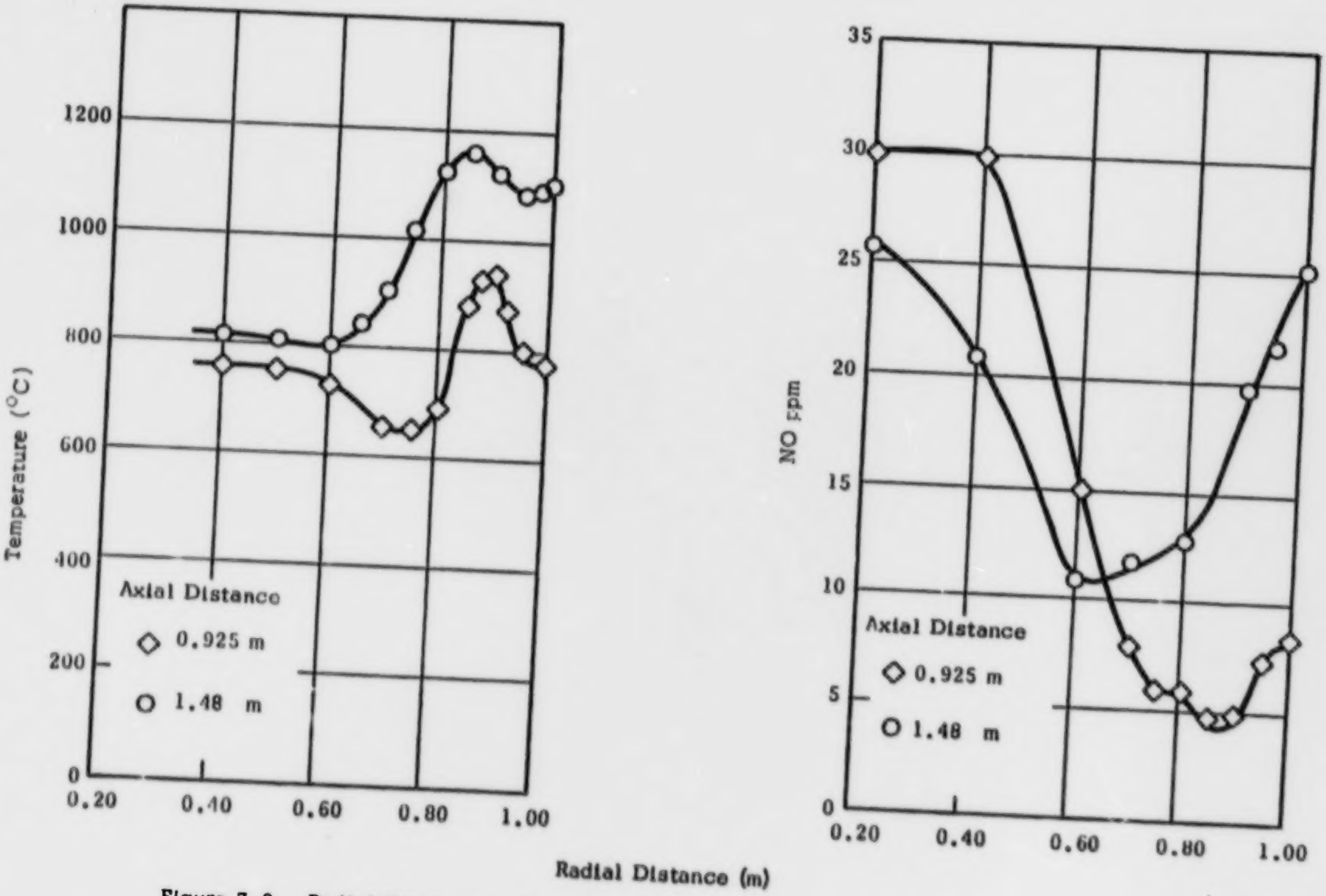

Figure 7.8 Radial Temperature And NO Distributions For Slightly Swirling Propane Flames (s=0.5)

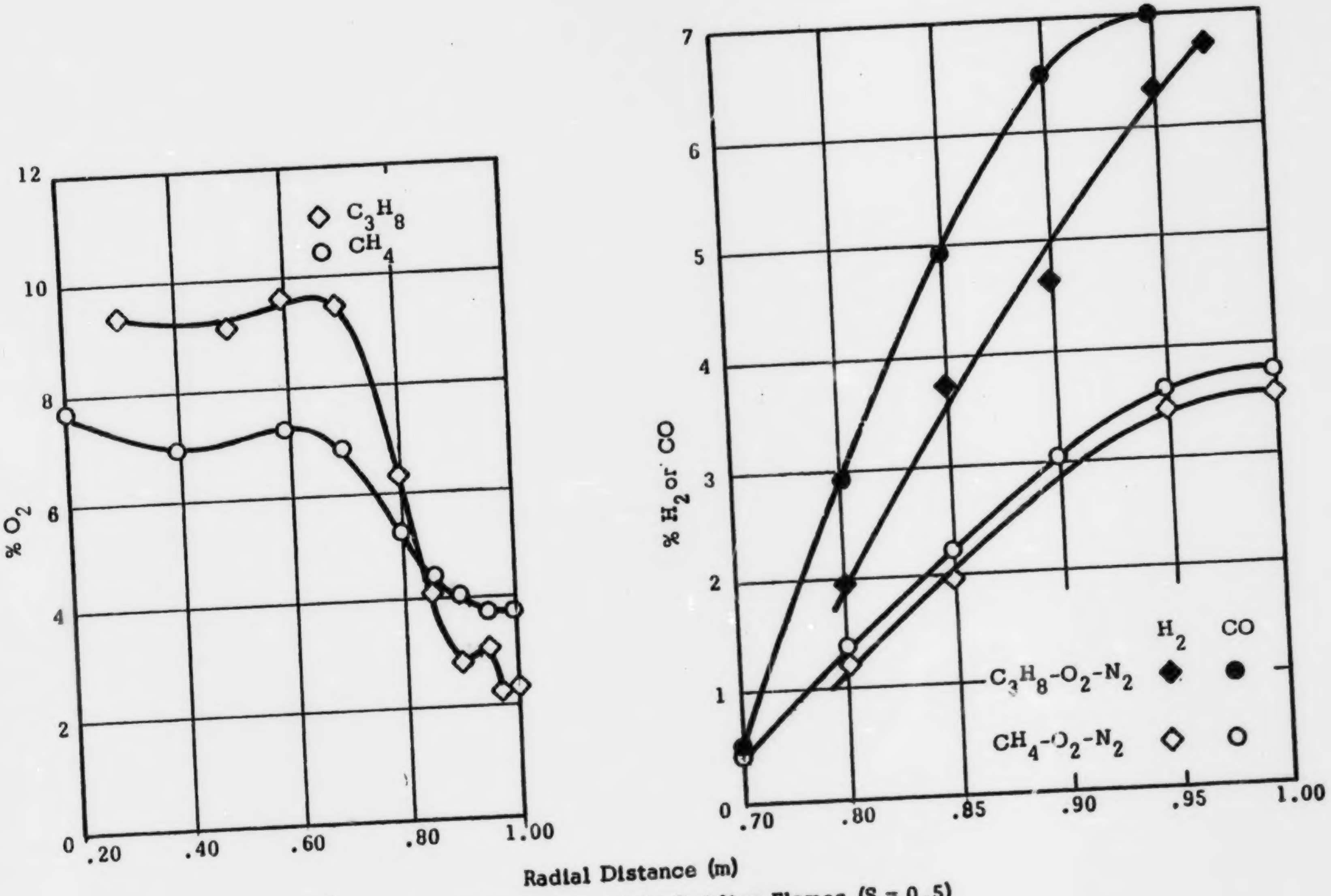

Figure 7.9 Radial Gas Concentrations For Slightly Swirling Flames (S = 0.5)

flames were real and the reasons for them.

The results presented in Fig. 7.10 show that for high intensity Type I flames an increase in swirl level causes an increase in the emission level. This behaviour is contrary to that of simple jet flames where emissions are reduced with increasing swirl. The influence of swirl on Type I flames can be explained by reference to Fig. 7.11 which is a diagramatic representation of the conditions prevailing at the bulbous root of the flame. The axial fuel jet penetrates the internal recirculation zone which forms an annulus around the fuel jet. Mass exchange takes place between the fuel jet and the reverse flow such that:

- the fuel jet entrains combustion products
- some of the fuel is stripped from the main jet and is returned to the root of the flame.

The hot fuel in the internal reverse flow region cannot react until it is mixed with air. Combustion takes place in the flame region which separates the zone of internal recirculation from the swirling air flow. The fuel remaining in the main jet reacts further downstream with air that has been diluted with inerts due to entrainment of external recirculation. If the swirl level is increased the intensity of the internal reverse flow increases and more fuel in entrained by the internally recirculating products. The formation of the internal recirculation marks the conversion of a simple jet flame to a type I. Measurements suggest that minimum emissions occur at this point. The increased emission with swirl for Type I flames is due to a redistribution of the zones of heat release. As the swirl increases more fuel is entrained by the internal recirculation and reacts at the base of the flame. Thus less heat release occurs in the main flame body. Heat release at the flame base will produce more NO because:

- T_F will be higher since dilution of the reactants will be less;
- the rate of decrease of T_F to T^1 will be slower because the internally recirculating gases have a higher temperature than externally recirculating gases.

Consequently, after initiation of the internal recirculation zone any further increase in swirl increases the emission of nitric oxide.

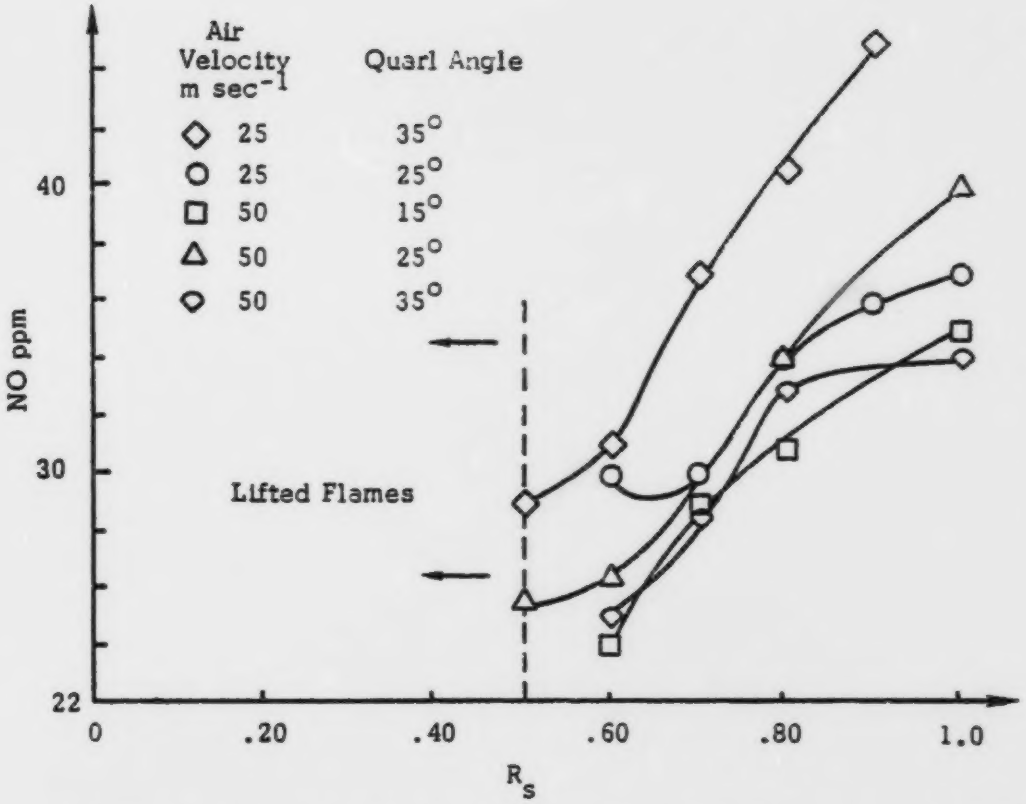

Figure 7.10 The Influence Of Swirl Level On NO Formation In High Intensity Type I Flame

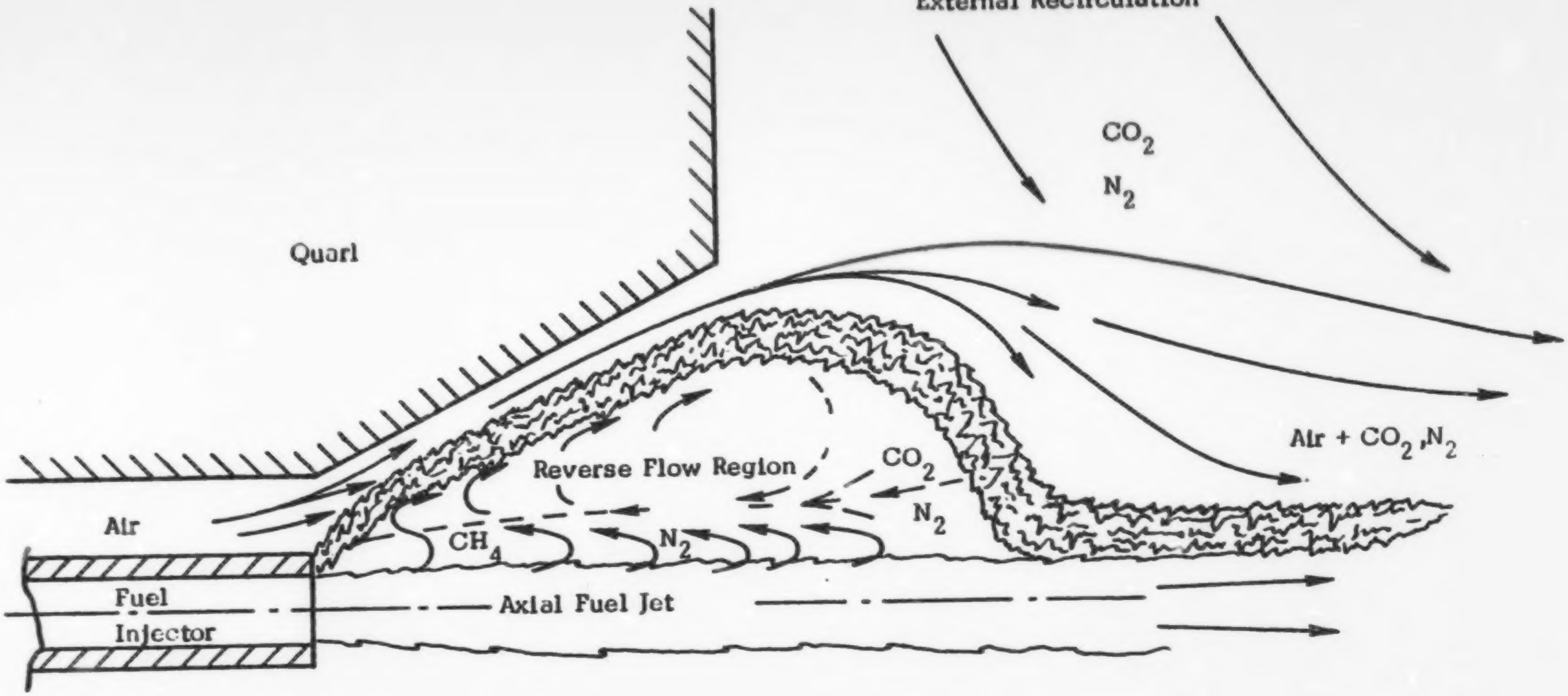

Figure 7.11 Diagramatic Representation Of Conditions At The Base Of A High Intensity Type I Flame

Three other burner parameters influence the emission of nitric oxide from high intensity Type I flames; the throat velocity, the angle of the burner divergent and the position of the fuel injector.

The velocity of the combustion air: the mass of external recirculation entrained by the flame jet is dependent upon the momentum of the combined primary and secondary jets. Thus increasing the velocity of the air supply increases the mass of external recirculation entrained by the flame jet. This will tend to reduce nitric oxide formation because the combustion air will contain a greater proportion of inert diluent and the flame products will be quenched more readily. The results presented in Fig. 7.10 show that the maximum emissions occur with the lower air velocity.

The angle of the burner divergent: the angle of the burner divergent has two effects upon flame characteristics. The smaller the angle the longer and narrower the flame. Also there is an optimum angle of approximately 25° for flame stability.

The increase in stability is believed to be associated with the strength of the internal reverse flow region. "Strength" is a nebulous term and may refer to either the reverse flow velocity or the mass flow rate of recirculating products. Under the circumstances discussed above it refers to the ability of the reverse flow to entrain fuel. The results presented in Fig. 7.10 suggest that with Type I flames the most important factor is the entrianment of external recirculation. At $25 \mathrm{m.sec.}^{-1}$ the narrower angle gives the minimum emission because the longer flame means that the combustion is delayed and the air will be more dilute thus reducing T_F. At high air velocities the angle of the quarl has a negligible effect because the entrainment of external recirculation is dominated by the higher jet momentum.

The position of the fuel injector: moving the fuel injector from the throat to the exit of the quarl means that less fuel is entrained by the internally recirculating products. Consequently the emission of nitric oxide from Type I is lower when the injector is at the exit rather than in the throat of the quarl. The emissions from lifted flames are generally low (See Fig. 7.1) because reaction takes place remote from the fuel injector. The total heat release occurs after significant dilution with entrained products. Thus potential T_F of each zone of heat release are low and consequently emissions tend to be low.

7.2.2 High Intensity Type II Flames

Three different fuel injector types were used during the investigation to produce high intensity Type II flames:

- a low velocity single hole injector;
- a multihole divergent injector;
- a multihole radial injector.

The single hole injector was not used extensively as it tended to produce asymmetric flames and the following discussion will be restricted to the emission characteristics of multihole injectors. The emission characteristics of the two injectors are presented separately in Figs. 7.12 and 7.13. Comparisons between the two injectors are possible by reference to Figs. 7.14 a and b.

The effect of burner variables on the emission characteristics of high intensity Type II flames can be summarized by:

- increasing swirl reduces emissions with divergent fuel injection;
- the influence of swirl is less pronounced with radial fuel injection and trends are not clear cut;
- at the lower air velocity emissions from both injector types are dependent of divergent angle;
- optimum burner conditions for minimum emissions with both injectors are high swirl -35° burner divergent at throat velocity of 50 $m.sec.^{-1}$. These burner conditions also give minimum emissions at zero swirl.

It is believed that the mass transfer within the burner divergent dominates the production of nitric oxide in Type II flames. Unfortunately it is extremely difficult to make accurate measurements within the divergent in combusting flow. Consequently, the discussion of the phenomena summarized above must be based upon a composite of the information presented in Section 3 , relevant isothermal measurements and intuition. The most significant influence of the fuel injector type upon emissions will be the variation in the fuel air mixing pattern.

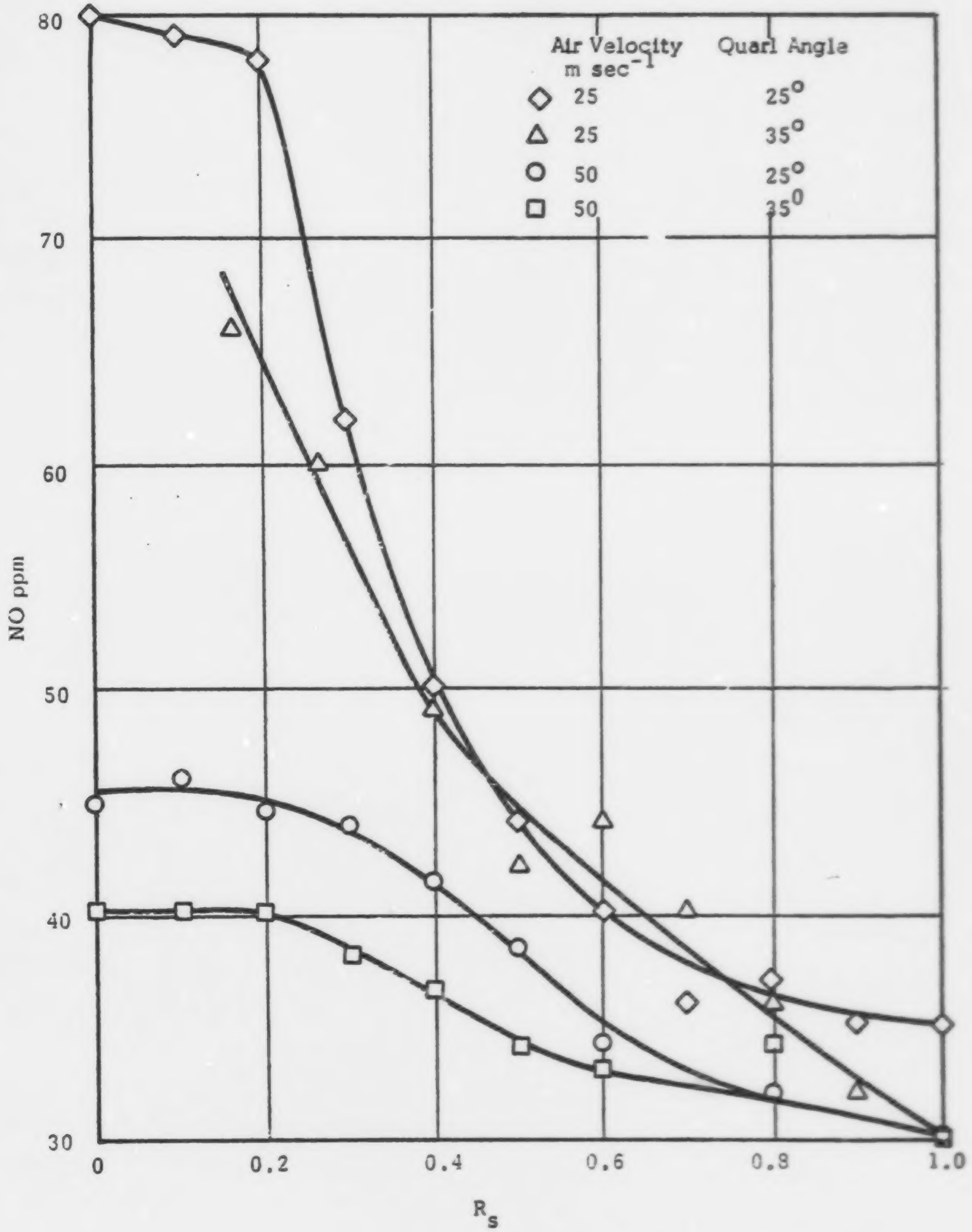

Figure 7.12 Emission Characteristics Of High Intensity Type II Flames Produced With A Divergent Fuel Injector

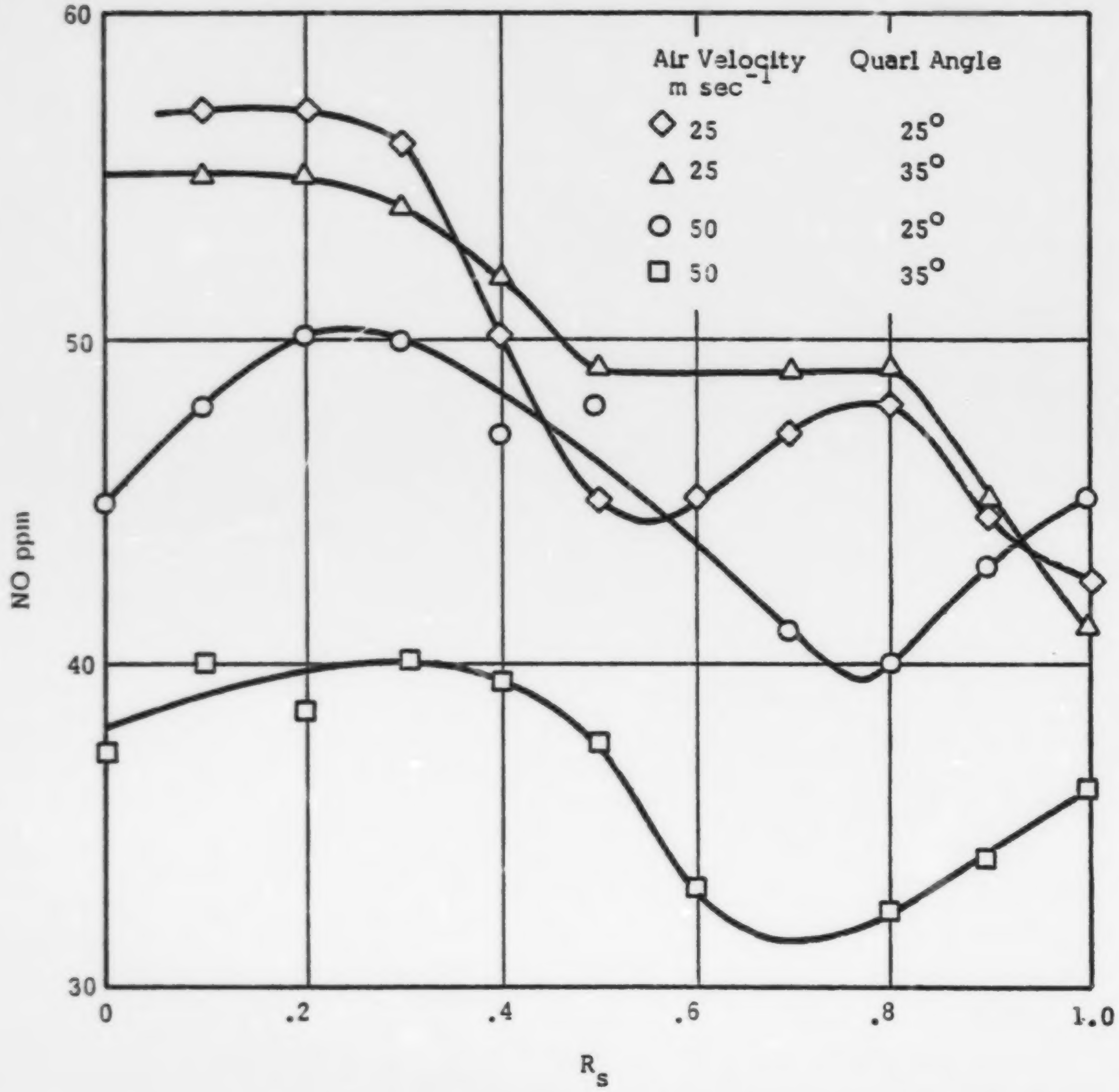

Figure 7.13 Emission Characteristics Of High Intensity Type II Flames Produced With A Radial Fuel Injector

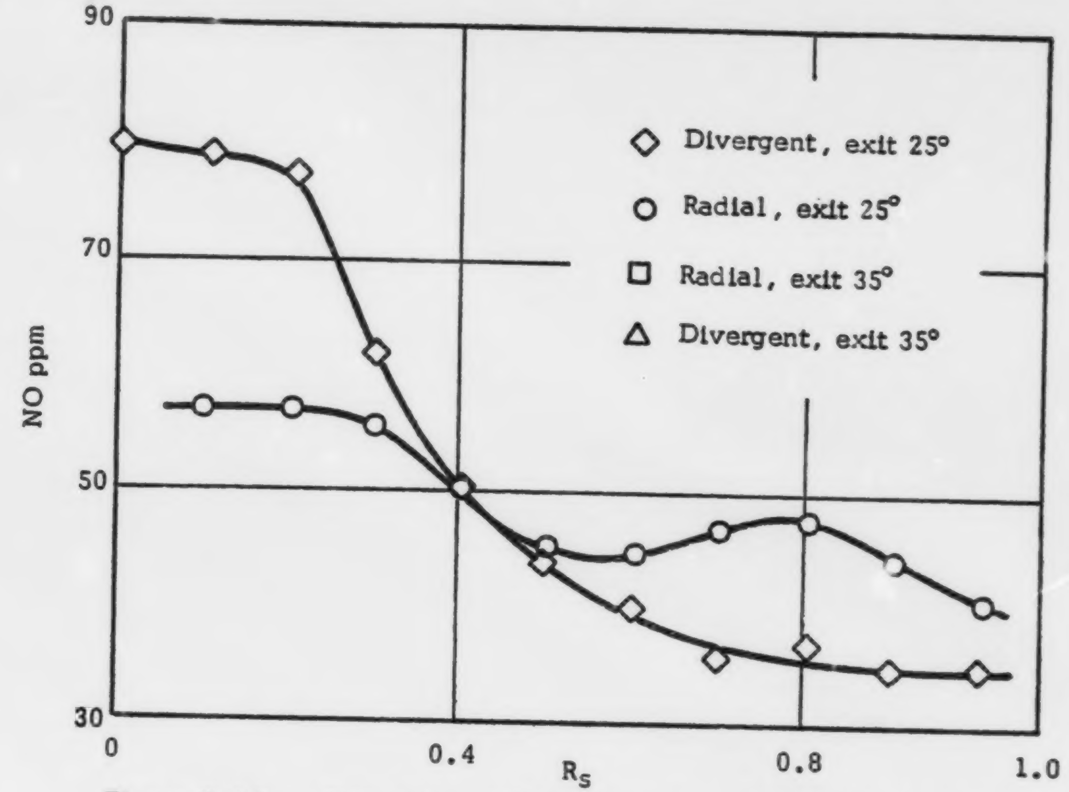

Figure 7.14a Comparison Of Emission Curves Multihole Injectors (25 m sec^{-1})

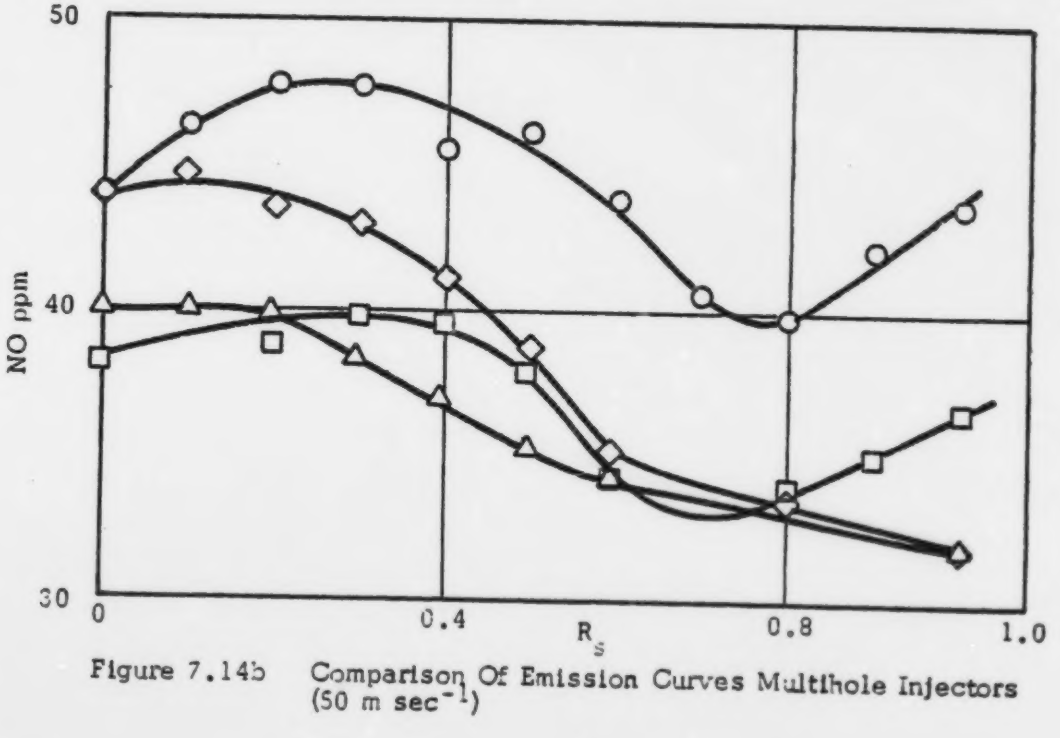

Figure 7.14b Comparison Of Emission Curves Multihole Injectors (50 m sec^{-1})

From the earlier discussion the fuel air mixing pattern will control the distribution of the total heat release between many separate zones each with its own equivalence ratio and degree of dilution. The burner variables will influence the size and intensity of the recirculation zone and the amount of entrained combustion products.

The radial fuel injector will produce more rapid fuel air mixing than the divergent injector. This effect has been confirmed by isothermal tracer measurements with scaled injectors (Fig. 7.15). A tracer was added to the primary stream and the decay of tracer concentration and flow boundaries were measured within the divergent. The concentration measurements presented in Fig. 7.15 confirm that at zero swirl primary secondary mixing is much faster with the radial injector than with the divergent injector. Increasing swirl improves the mixing with both injactor types. With the radial injector the fuel is injected perpendicularly to the air stream. Therefore mixing of some of the fuel is most likely to take place without reaction taking place. Under these circumstances the total heat will be released over a wide range of equivalence ratios with consequent reduction of T_F. The divergent injector directs the fuel parallel to the air stream and mixing will take place at the boundary between the two, allowing more of the fuel to be burned in zones with an equivalence ratio close to unity and T_F will be higher. Thus at zero swirl and low velocities the emission from the divergent injector will be higher than that from the radial injector.

The results presented in Fig. 7.14a indicate that swirl has a greater effect upon emission from divergent rather than from radial injectors. The difference in the flow patterns within the divergent probably account for this observation. With the divergent injector the fuel and air can more easily be diluted with recirculating products prior to reaction than with the radial injector. Fuel air mixing will take place prior to reaction with the radial injector and significant contact with hot recirculating products will cause ignition of an undiluted mixture. Increasing swirl will increase the rate of temperature decrease of the high temperature products for both injectors.

Increasing the throat velocity decreased the emission levels produced with the

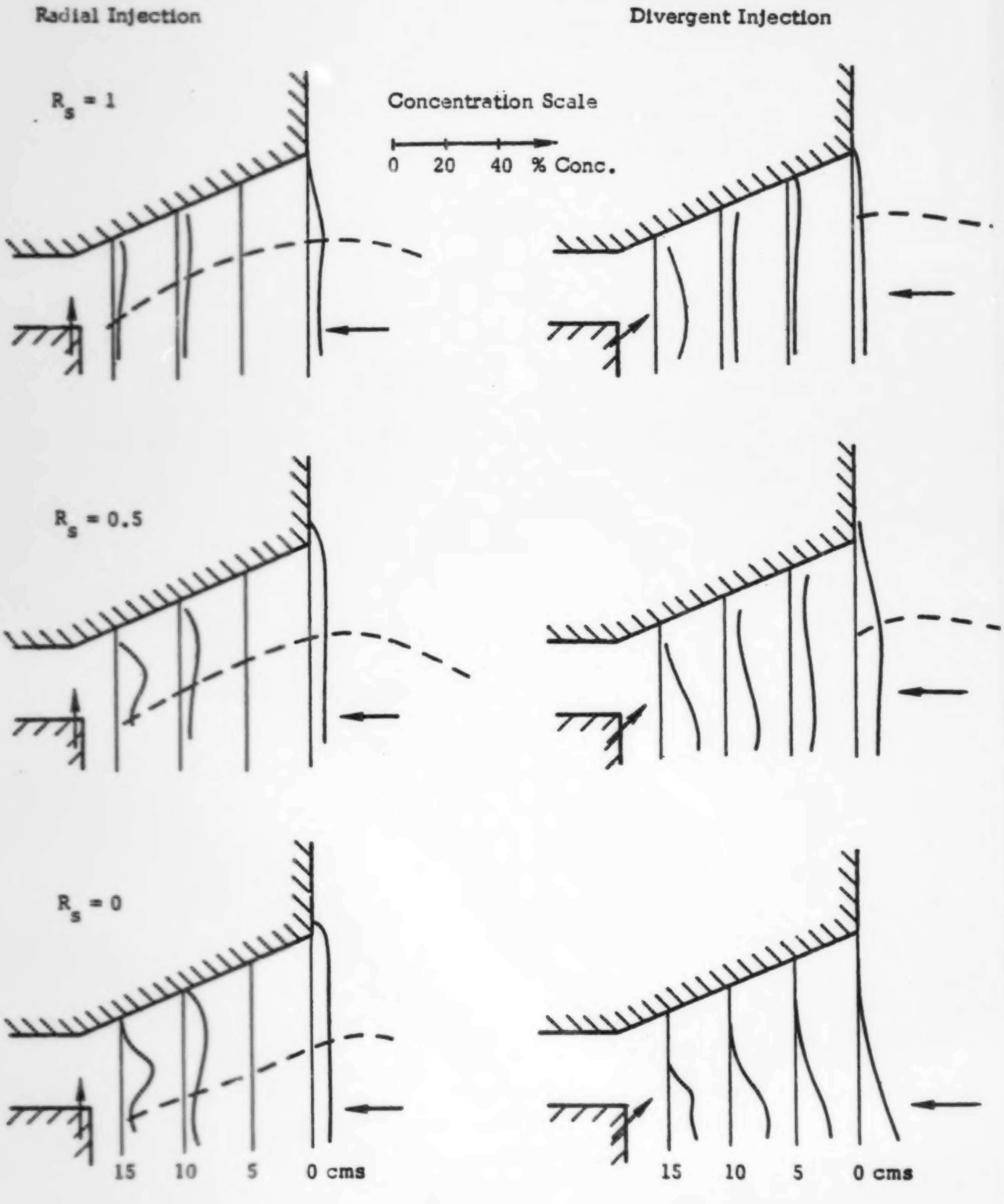

Figure 7.15 Isothermal Tracer Concentration Measurements Comparing Divergent And Radial Injectors

divergent injector markedly at low swirl levels. The emission levels measured with throat velocities of 50 m. sec.$^{-1}$ were similar for both multihole injectors. The formation of nitric oxide will be reduced at high throat velocities because of the increase rate of entrainment due to the higher jet momentum. It is not readily apparent why the burner divergent only has a significant influence upon the emission levels with high throat velocities.

For all the burner conditions investigated similar emission characteristics were observed when the radial injector was placed at the exit of the divergent (See Fig. 7.16). As the swirl level was increased the emission increased to a maximum and then decreased. The form of the emission curve is independent of burner divergent and throat velocity. In an attempt to explain these emission characteristics conditions at the injector can be represented as in the sketch shown in Fig. 7.17. At zero swirl the internal reverse flow zone is formed in the wake of the fuel injector. Mixing between the air and fuel is not so efficient as when the injector is in the throat due to a decrease of air velocity because of expansion in the divergent. Combustion is delayed, thus more of the air becomes diluted with inerts and T_F is reduced and emissions are low. When the swirl is increased the fuel and air mix faster before significant dilution takes place, T_F increases as does the emission. As the swirl is increased the internal reverse flow zone is stabilized and then it begins to increase in size. The boundary of the reverse flow zone moves upstream until it covers the fuel injector (See Fig. 7.17). At this point three factors have a major influence upon nitric oxide formation:

- improved fuel air mixing produce earlier combustion before dilution thus NO increases;
- the fuel jet must now penetrate the reverse flow zone, entrain inert products thus NO formation decreases;
- increased swirl causes an increase in entrainment and NO formation is reduced.

Considering the above factors it can be seen how the emission can increase and reach a maximum and then decrease.

Evidence in support of the above explanation has also been obtained by tracer studies in isothermal flow. The change in position of the reverse flow zone with increasing swirl can be seen in the results presented in Fig. 7.18. It can also be

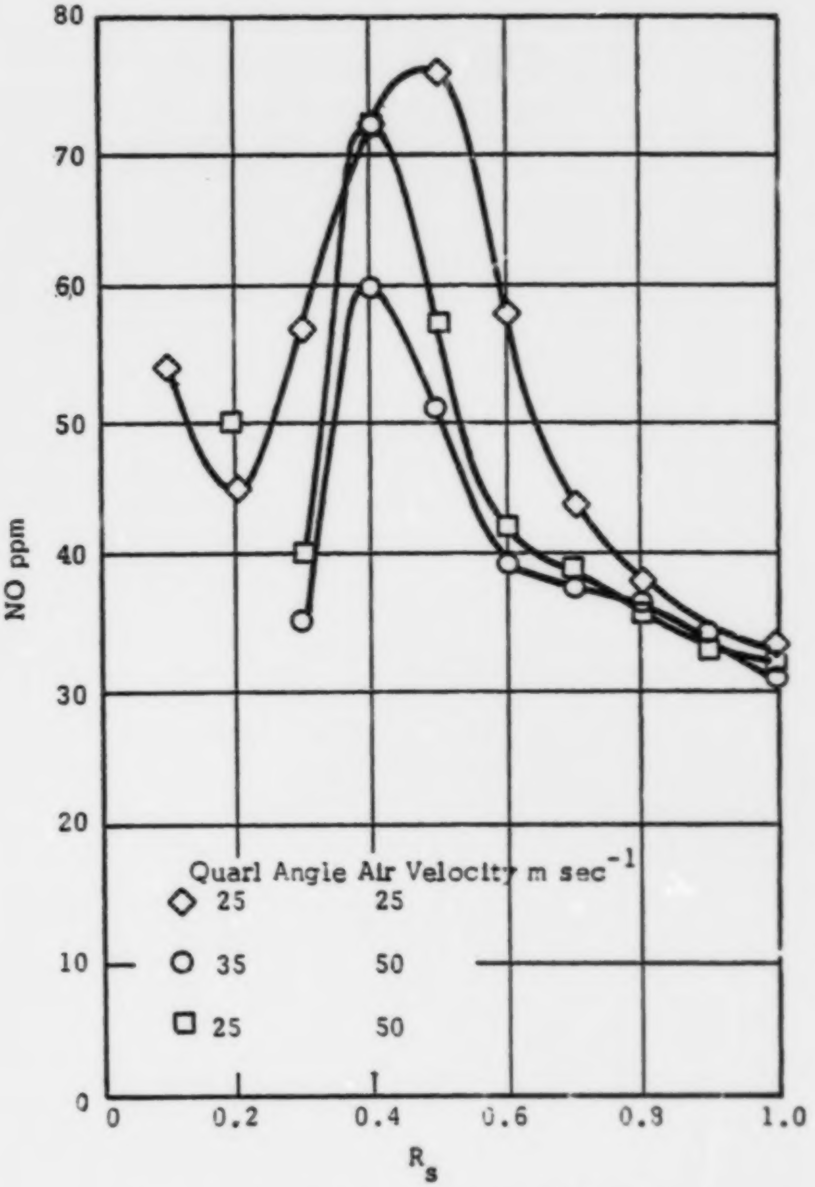

Figure 7.16 Emission Curves For Type II Flames Produced With A Radial Injector At The Burner Exit

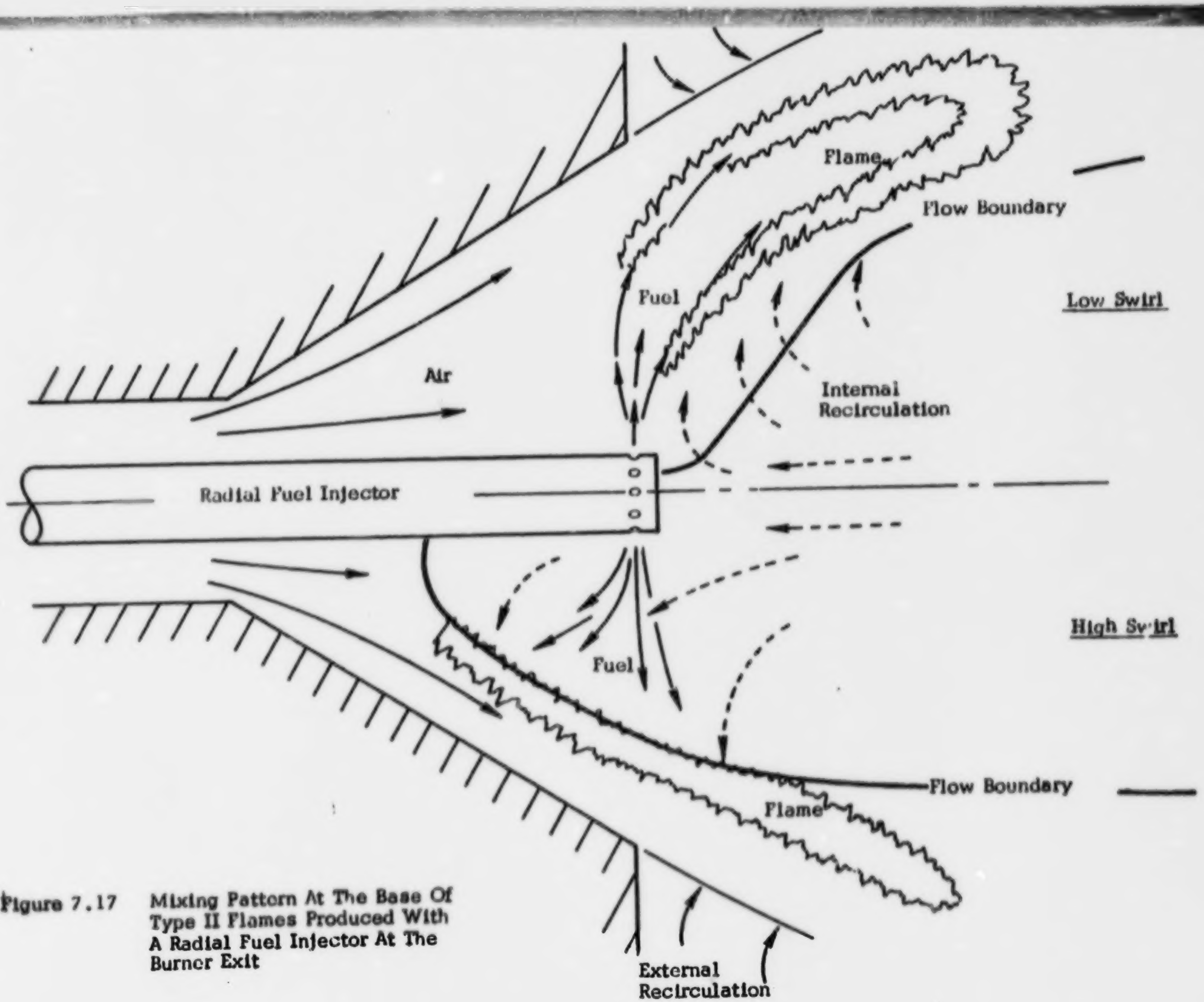

Figure 7.17 Mixing Pattern At The Base Of Type II Flames Produced With A Radial Fuel Injector At The Burner Exit

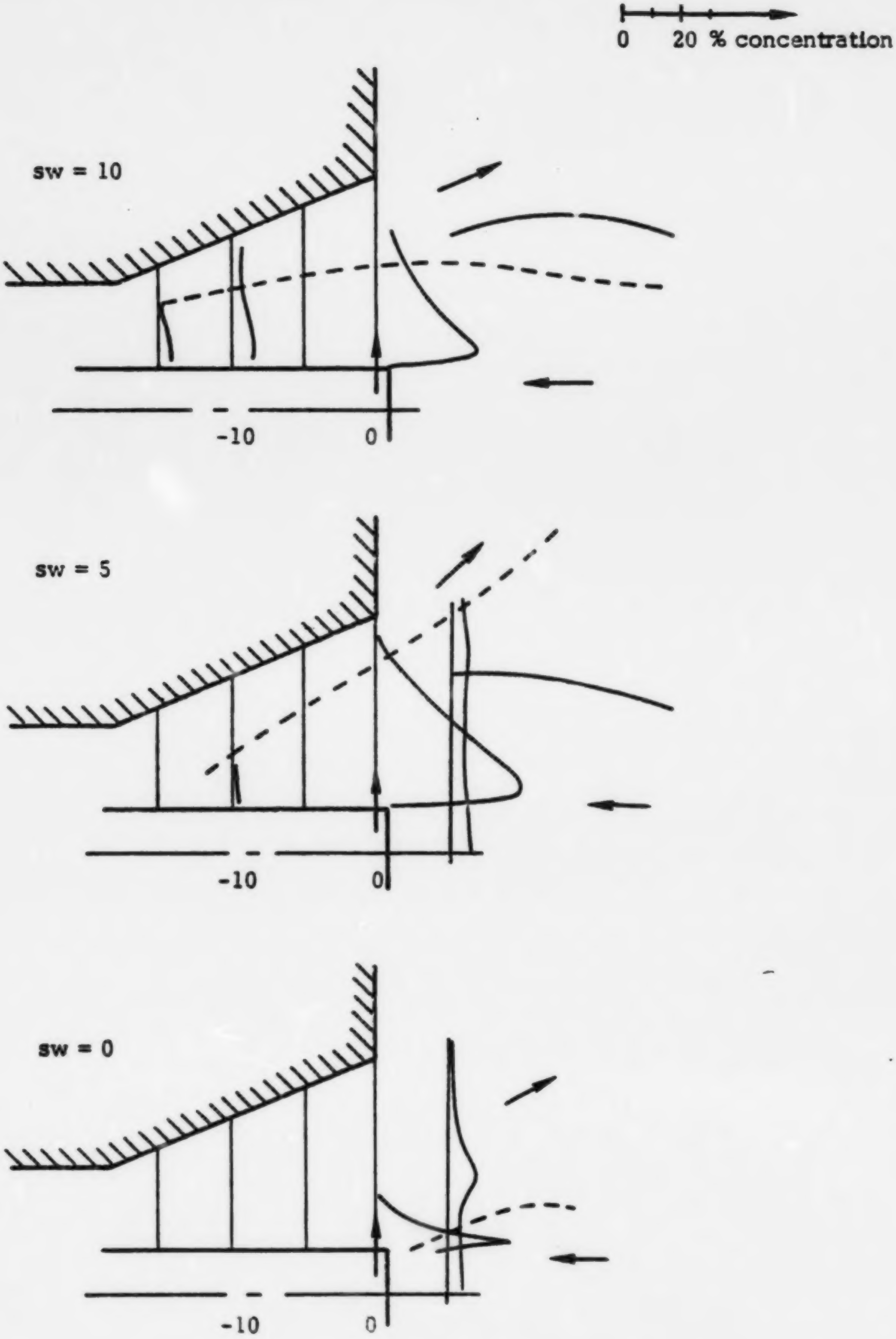

Figure 7.18 Primary Concentration And Position Of Recirculation Zone For A Radial Injector Placed At The Exit Of The Burner

seen that the primary matter is carried upstream by the reverse flow allowing combustion to take place within the divergent.

7.2.3 The Influence of Flame Quench Rate

Considering the dependence of nitric oxide formation in natural gas flames upon temperature it is not surprising that quench rate has a significant influence upon nitric oxide emissions. Two different experiments were carried out which illustrate this influence:

- refractory burner divergents were used instead of water-cooled divergents;
- the furnace cooling load was reduced.

For the three injector types tested emissions were lower when a water-cooled divergent was used rather than a refractory one. In all three instances the form of the emission curve is similar (See Fig. 7.19). The surprising feature of the results is the relative influence of the divergent type with the three fuels. The high velocity single hole injector which produced a high intensity Type I flame showed the most influence.

When the furnace cooling load is reduced, the bulk gas temperature increases thus the enthalpy of the reactant mixture is increased and the rate of temperature decrease from T_F to T^1 is reduced. The influence of furnace cooling rate on NO formation is also dependent upon flame type. It can be seen from Fig. 7.20 that the bulk gas temperature affects emissions from Type I flames more than from Type II flames. In Type II flames mass exchange between the flame volume and the internal recirculation zone predominates. The temperature of the internal recirculating gases is not strongly dependent upon furnace cooling load. Therefore the bulk gas temperature has less influence upon nitric oxide formation than in Type I flames which are longer and the volume entrains larger masses of external recirculation.

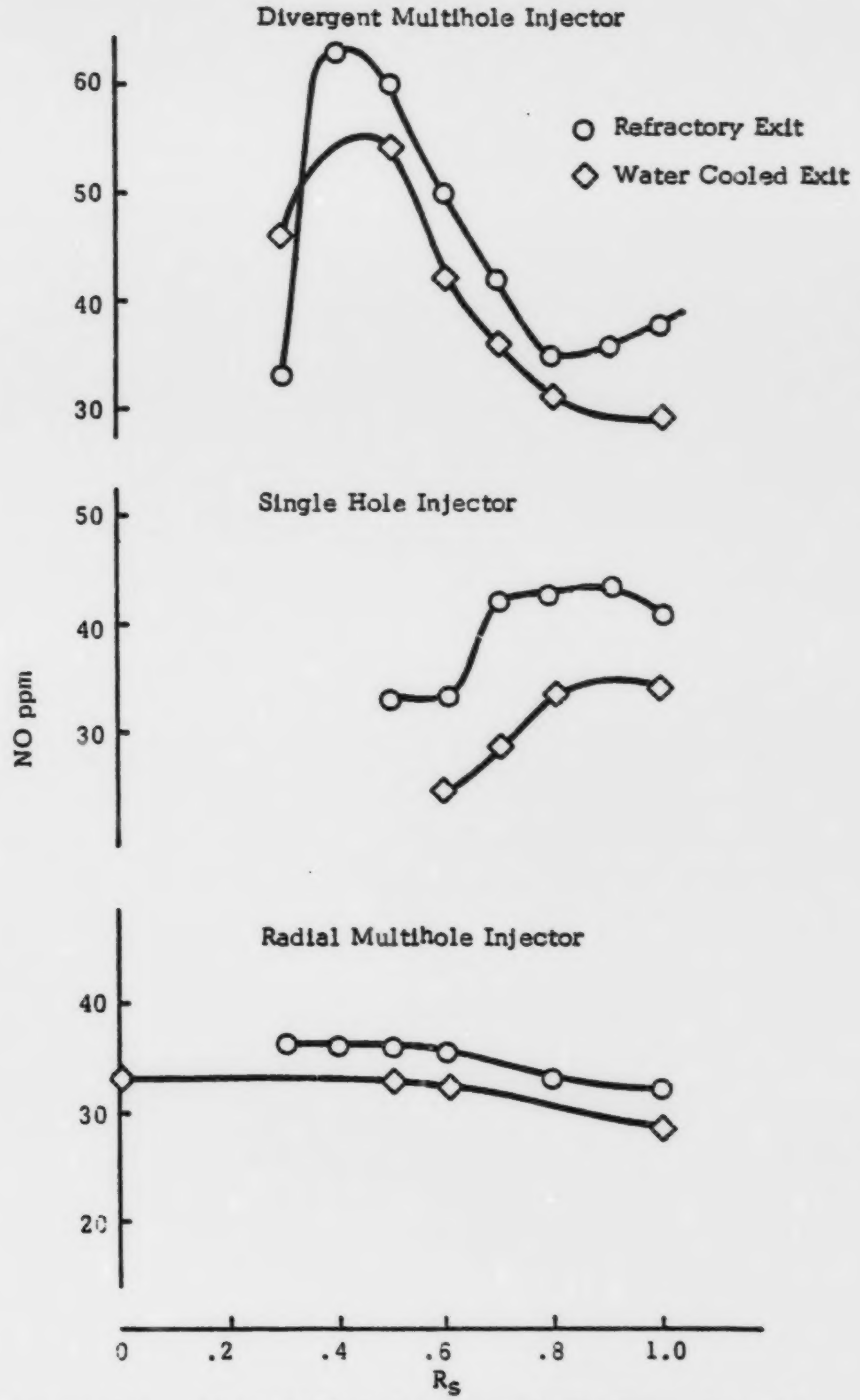

Figure 7.19 Effect Of Heat Extraction At The Flame Base On NO Emission

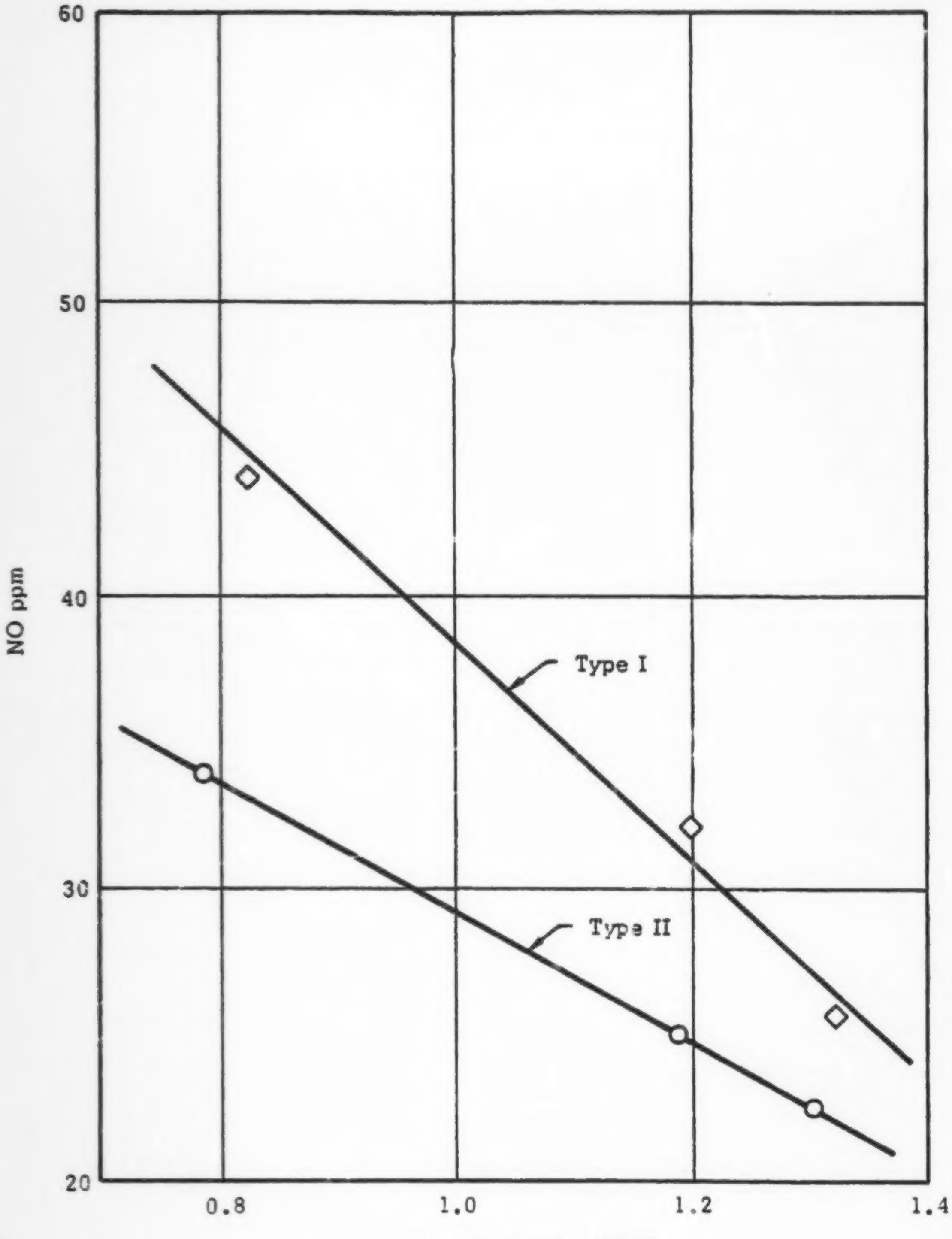

Figure 7.20 Influence Of The Heat Extraction On The NO Formation In Type I And Type Flames

7.3 Nitrogen Dioxide Formation in Natural Gas Flames

It is generally accepted that the oxide of nitrogen produced initially in combustion processes is nitric oxide (NO).

During the natural gas investigations significant quantities of nitrogen dioxide (NO_2) were found in the flue gases. Figures 7.21 and 7.22 present flue gas (NO, NO_2 and NO_x concentrations at various swirl levels for a Type I flame and a Type II flame. It can be seen that although the NO_x emission varies significantly the NO_2 content remains constant with swirl.

The oxides of nitrogen concentrations were determined with a chemiluminescent analyzer. The NO_x concentration was determined by diverting the sample through a heated packed quartz tube to thermally convert NO_2 to NO prior to analysis. The NO_2 concentration is then evaluated as the difference between the NO_x and NO concentrations. The temperature of the flue gases was greater than 1000°C and in the first instance it was thought that the nitrogen dioxide was being produced at low temperatures by the oxidation of nitric oxide within the sampling system. However, even when the flame was switched off and nitric oxide added to the air supply into the hot furnace nitrogen dioxide could not be detected in the flue gas.

Detailed flame measurements enable an explanation to be given which accounts for the presence of NO_2 in the high temperature flue gases. Fig. 7.23 shows radial profiles of temperature, oxygen, NO and NO_2 concentrations 10 cm from the burner exit for a Type II flame. The nitric oxide concentration has a high value (110 ppm) in the external recirculation which drops almost to zero when it is mixed with the air jet. The peak of oxygen concentration corresponds to the minimum of the nitric oxide curve. The nitric oxide concentration increases to a maximum in the internal recirculation zone. The nitrogen dioxide has a low value in the external recirculation (7 ppm) but the concentration increases as the oxygen concentration increases. Concentrations of NO_2 greater than 35 ppm were measured and the nitric oxide concentration at that point was 17 ppm. Unfortunately the measuring system is not capable of measuring NO_2 concentrations in samples containing CO, H_2CH_4 etc.

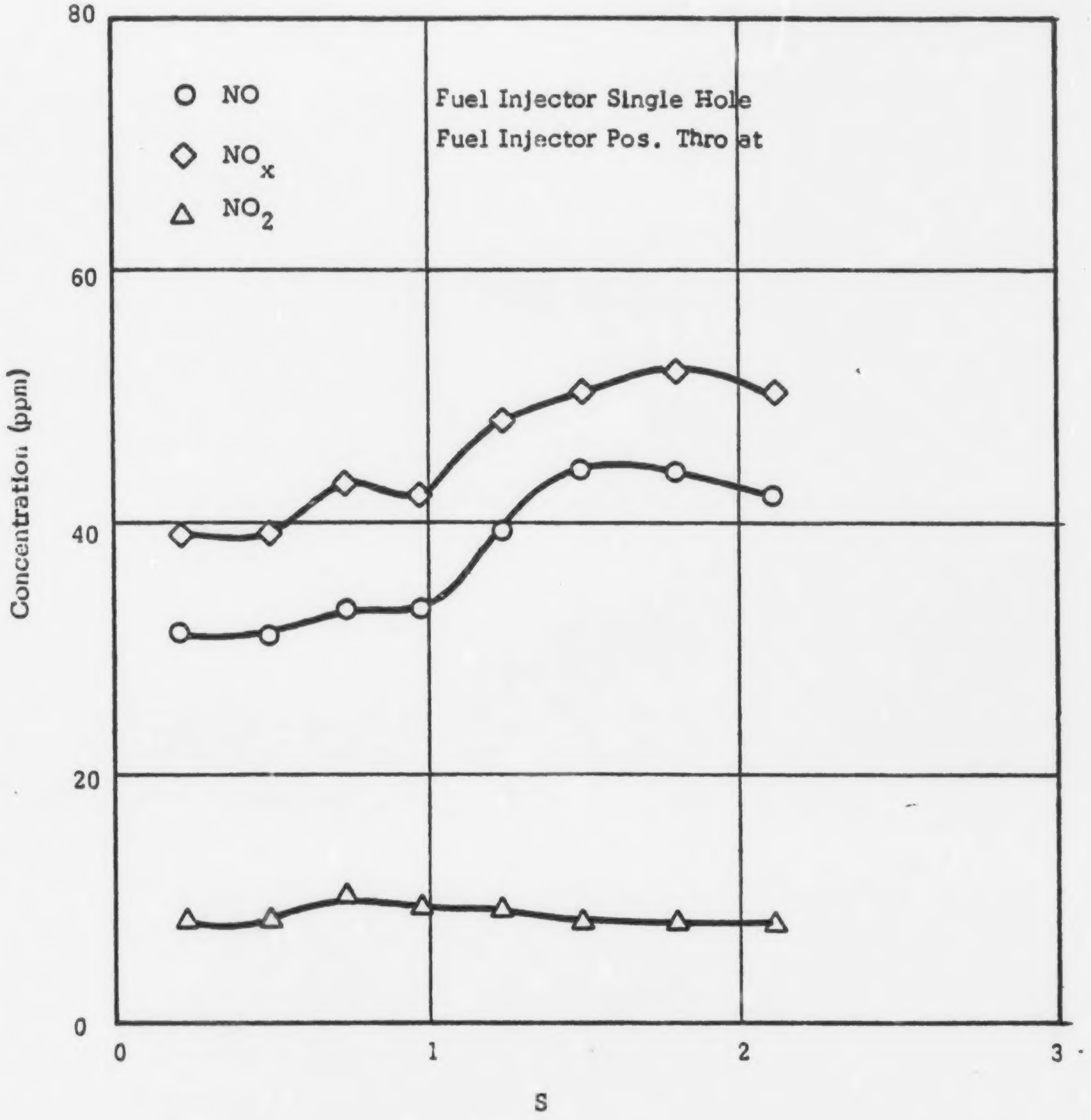

Figure 7.21 Flue Gas Concentration Of NO, NO_x and NO_2 As A Function Of Swirl Number For A High Intensity Type I Flame

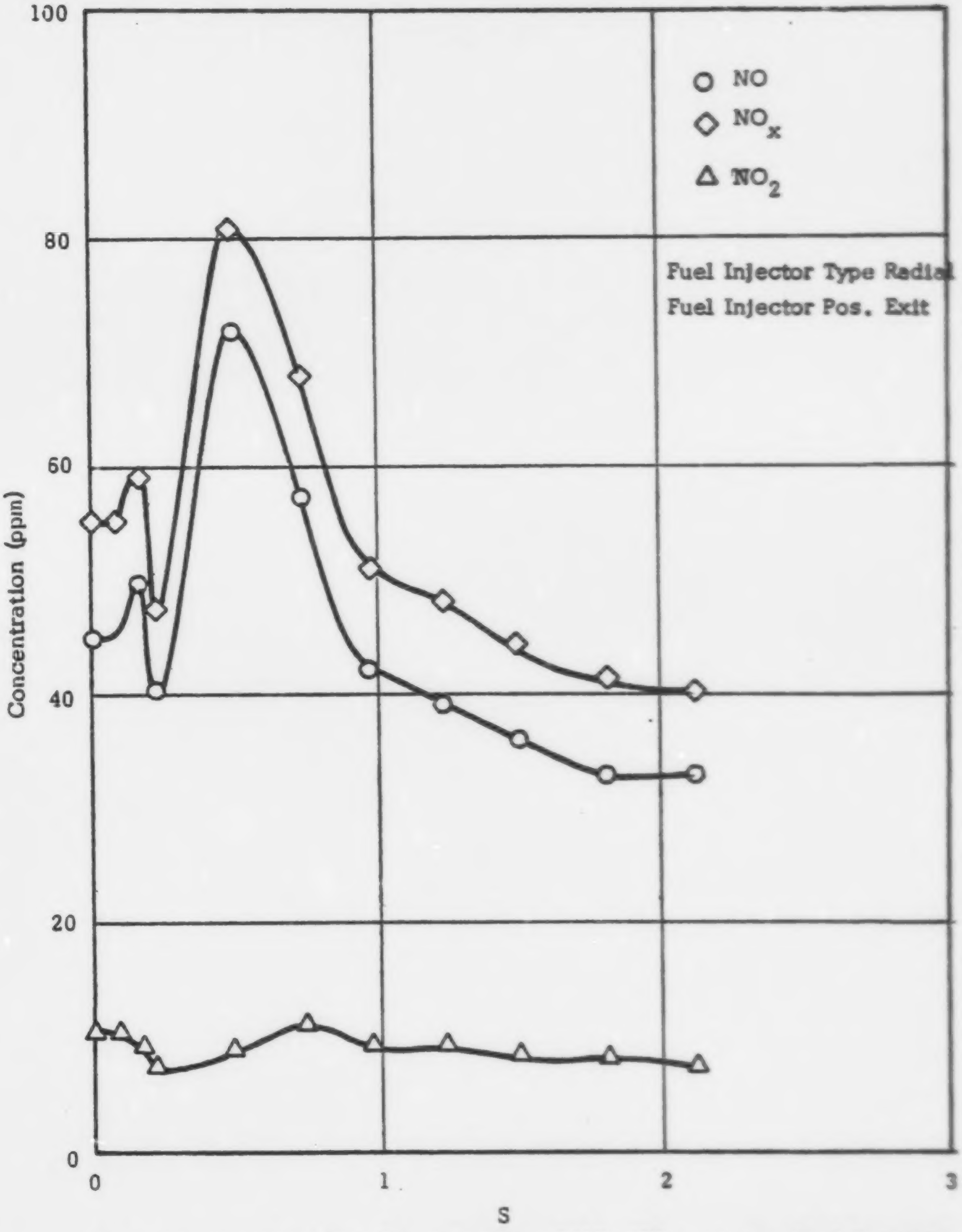

Figure 7.22 Flue Gas Concentration of NO, NO_x and NO_2 As A Function Of Swirl Number For A High Intensity Type II Flame

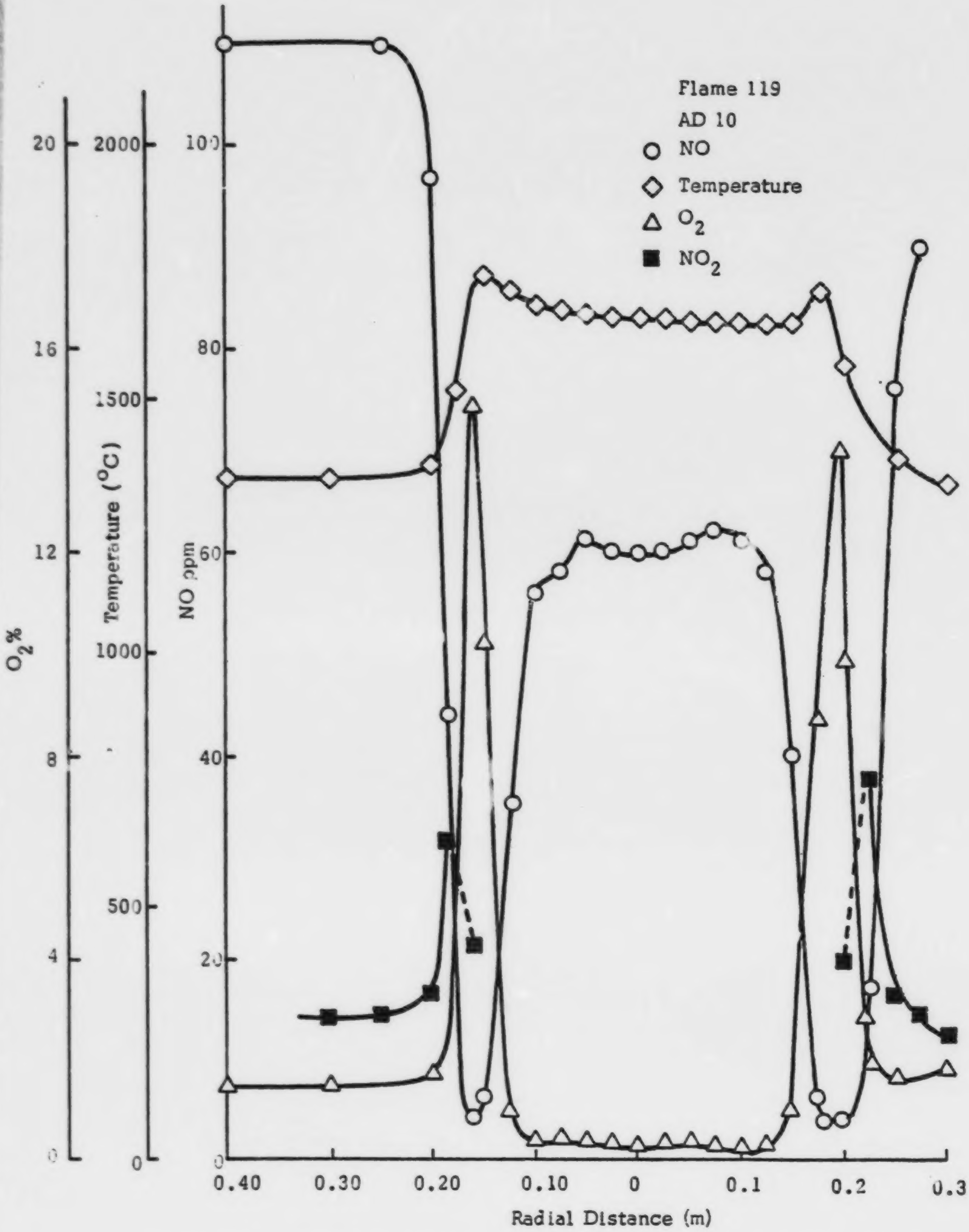

Figure 7.23 Radial Distributions Of Temperature O_2 NO And NO_2 10cm From The Burner Exit In A Type II Natural Gas Flame

The reduction in NO_2 concentration to zero in the flame region may not be a real effect but due to the reduction of NO in the converter by CO, H_2 and solid carbon. However, the increase of NO_2 concentration is a real effect and cannot be attributed to the converter. Thus it appears that there may be considerable NO_2 concentrations in the flame region. There are three explanations for the observations concerned with nitrogen dioxide:

- nitric oxide in the recirculation is oxidized in the air jet;
- nitrogen dioxide is produced in the flame region;
- the compound is not nitrogen dioxide but is something which can be converted to nitric oxide within the quartz converter.

Nitrogen dioxide is unstable at high temperatures and it will decompose as the combustion products flow along the furnace. This effect is illustrated in Fig. 7.24. Plug flow conditions can be considered to be established 2 m from the firing wall under highly swirling conditions. After this point it can be seen that although the NO_x content of the combustion products remains constant the NO level increases and the NO_2 concentration decreases. This effect may explain observations reported by other investigators (7.2) and also at the IFRF where combustion is completed, the bulk temperature is low and dropping and yet nitric oxide concentrations are increasing.

7.4 Correlation of Emission Data

Wendt (7.3) has investigated the use of dimensional analysis to model the effect of swirl on NO_x emission levels from a test furnace. He concluded that the conventional dimensionless analysis approach was not very useful but that considerable insight into the problem could be gained from what Spalding (7.4) refers to as partial modeling. Wendt suggests that since nitric oxide formation in natural gas flames is a function of the furnace, the input conditions and the burner variables should be included. Although the populations are small, an attempt has been made to correlate the emission data against a number of parameters related to the burner variables.

Table 7.1 summarizes all the experimental data obtained at a fixed thermal input, furnace cooling load and excess air.

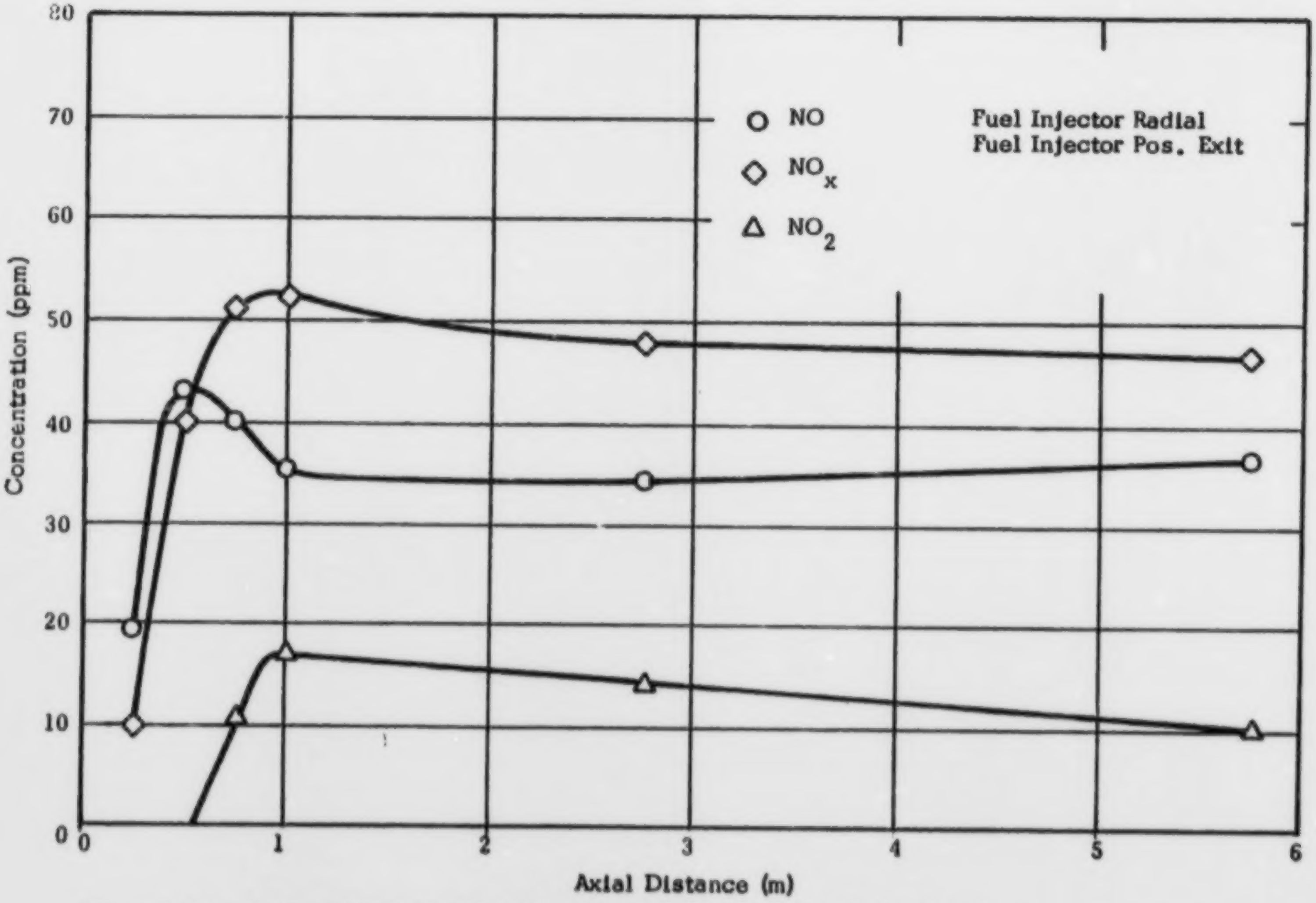

Figure 7.24 Axial Distribution Of NO, NO_2 And NO_x In Natural Gas Flame

TABLE 7.1 Summary of Experimental Data

Injector Type	No. of Observations	Mean Value ppm	St. deviation	Max. value ppm	Min. Value
Single hole	42	36.2	6.02	50	23
Divergent multihole	53	42.6	11.2	80	30
Radial multihole	69	45.6	9.93	76	28.5
All types	164	41.6	10.5	80	23

Multiple linear regression analysis was used in an attempt to correlate the data against the experimental variables. The results are presented in Table 7.2 and as could be expected from the emission curves presented earlier the correlations are not very significant.

Table 7.2 Multiple Linear Regression Correlations

Injector Type	Regression Equation	Correl. Coeff.	Standard Deviation
Single hole	ppm NO= 42- .231 (Vel.m.sec^{-1})	0.466	5.4
Divergent multihole	ppm NO= 89-13.24 (swirl number) - 6.48 (vel.m.sec^{-1}) - 3. 85 (quarl angle)	0.785	7.1
Radial multihole	ppm NO= 69, 2-8. 8 (swirl number) -441 (vel.m.sec^{-1})	0.664	7.54
All Types	ppm NO= 61, 9-8. 21 (swirl number) -.357 (vel.m.sec^{-1})	0.542	8.91

Further correlations were attempted with 25 "manufactured " parameters which are listed in Table 7.3. The correlation coefficient for all the 25 variables was 0.93 and the standard deviation was 6.4 ppm. After rejection of all variables with less than 1% significance the correlation coefficient was 0.81 and the standard deviation 6.5 ppm. Factor analysis of the 25 variables suggests that there are five independent aspects. However, the meaning of some of them is obscure. Aspect I is obviously concerned with swirl, aspect IV is related to the burner geometry and aspect III is concerned with the effect of the fuel injections angle. One encouraging feature of the analysis was a high single correlation between the variable $G_p U \cos\varphi (G_s V [1 + S^2])^{-1}$ and [NO] of 0.59. This variable attempts to express the relationship between the swirling kinetic energy and the kinetic energy in the fuel jet. The theoretical pressure energy required to produce solid body rotation in an annular air jet is given by:

$$P = \frac{1}{2} V^2 \left[1 + \frac{2R_o^2}{(R_o^2 + R_1^2)} S^2 \right] \tag{22}$$

This expression is superior to swirl number as a means of correlating emission data from divergent injectors. However, except for this case, the swirl number appears to be the best means for correlation of NO emissions where the tangential motion is produced by similar swirl generation devices.

TABLE 7.3 PARAMETERS USED IN MULTI-REGRESSION ANALYSIS

Variable	greater than 1% significance	aspect
$S^{\frac{1}{2}}$		I
S	x	I
S^2		I
$\sin \varphi$	x	III
$\cos \varphi$	x	III
$G_p . G_s{}^{-1}$	x	II
$(G_p U) . G_s V^{-1})$	x	II
$(2l \tan \alpha + D)^2 . D^{-2}$		IV
$d^2 . (2l \tan x + D^2)^{-1}$		
$G_p U \sin \varphi . (G_s V [1 + S^2])^{-1}$		
$G_p U \cos \varphi . (G_s V [1 + S^2])^{-1}$	x	
V	x	
$S . D$		I
$S . D^{-1}$	x	I
$S^2 \sin \varphi$		
$(L - 1)L^{-1} \cos \frac{1}{2} \alpha S^{\frac{1}{2}} \sin \varphi$		V
$G_p . S . (1 + \cos \varphi)$	x	
α		IV
$[G_p . (G_s)^{-1}]^1 + \sin \varphi$		II
$G_s/G_p . S$	x	
$S^2 G_s V . (G_p U)^{-1}$	x	II
$(L - 1)L^{-1} \cos \frac{1}{2} \alpha . G_s(G_p)^{-1}$	x	V
$(L - 1)L^{-1} \cos \frac{1}{2} \alpha . G_s V(G_p U)^{-1}$		V
α^1		V
$G_s V (1 + S^2)$	x	

NOMENCLATURE

A - recirculation constant

c_p - specific heat

D - throat diameter

F - empirical heat release factor

G - momentum

k - emission, attenuation coefficient

L - length of quarl

l - distance of injector from the throat

N - exponent on the oxygen concentration

N - degree of oxidation

R - radius

S - swirl number

T - temperature

t - time

U - velocity of the primary jet

V - velocity of the secondary jet

V - volume of reactor

ρ - density

α - quarl angle

φ - injection angle

SUBSCRIPTS

p - primary

pf - plug flow

s - secondary

ws - well stirred

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8. DISCUSSION OF RESULTS – PULVERIZED FUEL FLAMES

The parametric investigations carried out with coal as a fuel had the same object as those using natural gas as a fuel – the isoluation of burner variables which are important to the formation of nitric oxide in coal flames.

8.1 Nitric Oxide Formation in Pulverized Fuel Flames

The combustion of pulverized coal is a complex process involving two-phase flow, rapid particle heating both by convection and radiation, devolatilization of the coal, combustion of volatile fractions and heterogeneous combustion of the solid char. During this process it is possible that both thermal and fuel NO can be produced. Consider the sketch of the lifted coal flame shown in Fig. 8.1 which separates the progress of combustion into three sections:

- the fuel preparation zone;
- the combustion of the volatile fractions; and
- char burnout.

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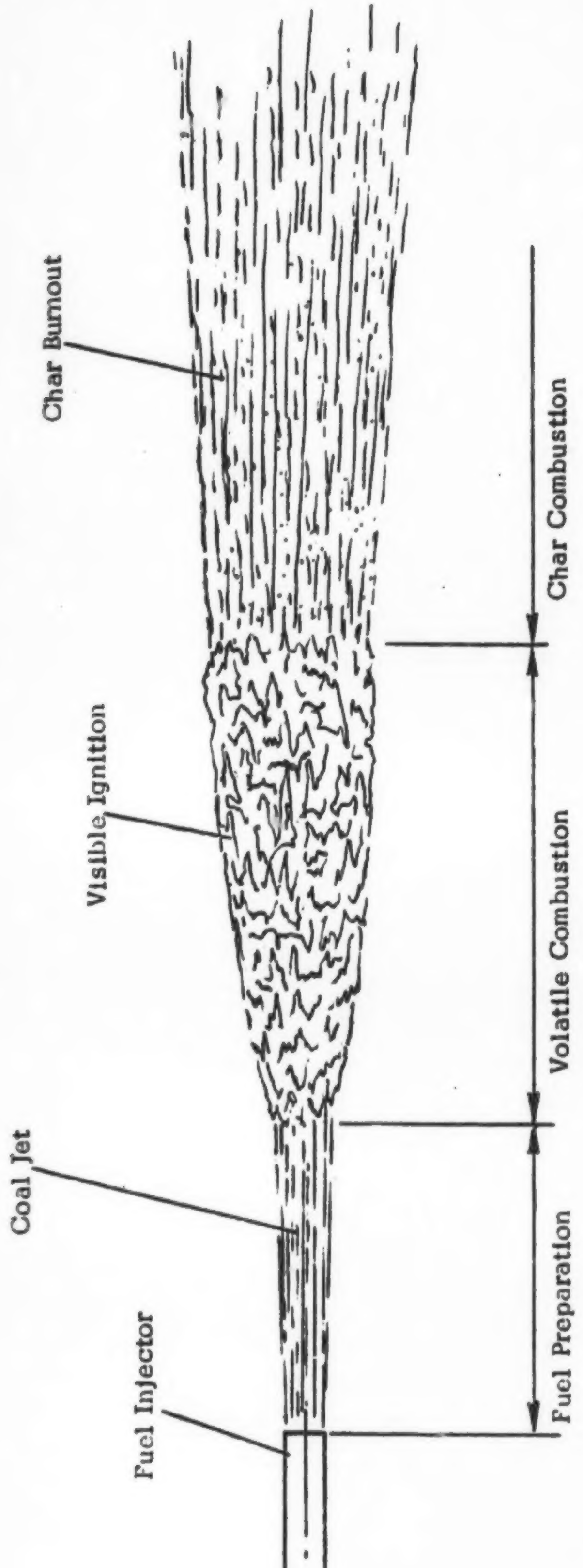

Figure 8.1 Idealized Sketch Of A Lifted Pulverized Coal Flame

8.1.1 Fuel Preparation

Pulverized fuel is coal that has been crushed and ground in order that it can be carried as a suspension in an air stream. The size distribution is extremely important for rapid burnout. The mean particle size is normally in the range 30 to 70 microns; however, inevitably there is a wide size range. Ten percent by weight of the fuel may be less than 10 microns and 10 percent greater than 100 microns. The fuel is injected into the furnace with air as a primary jet where the particles are heated and the ignition of the volatile fractions marks the end of the fuel preparation zone.

The primary jet normally contains 20 percent of the total air flow with a temperature less than 100°C. The secondary air is normally preheated to temperatures about 250°C. Some distance from the injection point the primary and secondary jets combine and the velocity distribution can be described by a single Gaussean distribution (8.1). The mixing rate between the two jets is dependent upon the ratio of the primary and secondary velocities and the thickness of the primary/secondary interface. The entrainment of hot external recirculation is dependent upon the combined mass and momentum fluxes.

In the fuel preparation zone the coal particles receive heat by two processes:

- by radiation from the furnace walls and the ignition front; and
- by convection from the preheated secondary stream and the entrained recirculating gases.

At a certain temperature the coal particles begin to decompose producing tars and gases collectively referred to as volatiles. The volatile fractions consist of carbon dioxide, water vapor, carbon monoxide, hydrogen, hydrocarbons, and nitrogen containing species. The composition of the volatiles and the weight loss of the particle depend upon the temperature/time history of the particle. Therefore, the rate of heating of the particle and its final temperature influence the quantity and composition of the evolved volatiles. The heating rate of p.f. particles normally exceeds 10^4 °C per second and the evolved gases are rapidly transported away from the particle surface.

8.1.2 Combustion of the Volatile Fractions

The volatile fractions will combust when they are mixed on a microscale with oxygen, the mixture lies within the flammable limits and a source of ignition is present. The distance between the visible ignition front and injector will be referred to as the ignition distance. This distance will be strongly dependent on the mixing rate of the coal with high temperature gases and will also be dependent upon furnace wall temperature. The combustion of the volatile coal fractions may be considered to occur in an envelope around the fuel jet or as a combustion zone associated either with a single particle or a group of a small number of particles.

When the flame envelopes the fuel jet,conditions such as those shown in Fig. 8.2 previal. The combustion region can be considered as a region into which fuel gases and oxygen pass from the fuel jet side and oxygen passes from the air side. Combustion products leave the region at both boundaries. The amount of oxygen mixed with the fuel gases will be dependent upon the initial amount of primary air and the rate of mixing between the primary and secondary streams prior to the ignition front. When combustion is associated with a single particle the volatile fractions can either burn in a diffusion flame around the particle or as a premixed flame in the wake of the particle.

8.1.3 Char Combustion

Char are those solid particles which remain after the devolatilization of the coal particles is complete. It may consist of soot particles, cenospheres and cellular particles. Sternling and Wendt (8.2) consider that the fuel nitrogen is divided between the volatile and char fractions of the coal. Analysis of the char remaining after proximate analysis of coal volatile content confirms that all the nitrogen is not volatile. This experiment can only be considered indicative and it will not give a quantitative value of the volatile fuel nitrogen for the pulverized coal particles since the time temperature history of the coal in the two cases is different.

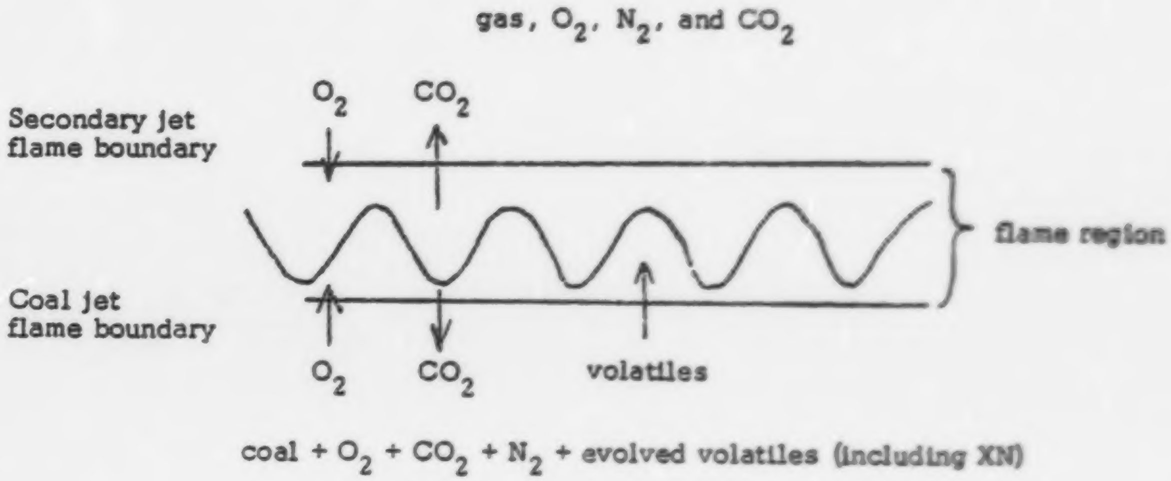
gas, O_2, N_2, and CO_2
O_2
CO_2
Secondary jet flame boundary
flame region
Coal jet flame boundary
O_2
CO_2
volatiles
coal + O_2 + CO_2 + N_2 + evolved volatiles (including XN)

Figure 8.2 Conditions At The Fuel Jet Boundary With An Injector Stabilized Flame

There is a fundamental difference between the combustion of the volatile and char fractions. The reactions involved in the combustion of the volatiles have finite rates but these are usually high compared with surface reactions involved in the combustion of the char. Also the combustion of the char particles involves several steps in sequence:

- transport of oxygen or other reactant gas to the particle surface;
- reaction with the surface;
- transport of the reactants away from the surface.

The overall reaction rate is of course dependent upon the slowest of these steps. It is probable that in the combustion of the char the product released at the surface of the particle is carbon monoxide (8.3). In Section 3.1 it was concluded that although the precise mechanism of fuel NO formation was unknown the amount produced could be attributed to two competing reactions:

$$I + R \longrightarrow NO + \dots \qquad (16)$$

$$I + NO \longrightarrow N_2 + \dots \qquad (17)$$

where I is an unknown nitrogen containing intermediate. Under fuel rich conditions reaction (17) is faster, but with fuel lean combustion large amounts of the fuel nitrogen can be converted to NO. Earlier it was stated that of the nitrogen found in the original coal a proportion can be considered to be "refractory" and remain in the char. It has been assumed that this fuel nitrogen does not form significant quantities of fuel NO. This assumption is based on the fact that the char particle will be surrounded by a stagnant boundary layer deficient in oxygen containing carbon monoxide. Thus even though NO were to be formed at the particle surface it could be reduced on passage through the fuel rich stagnant boundary layer.

Consequently, if the assumptions stated earlier are valid the influence of burner variables on nitric oxide formation in pulverized fuel flames can be explained by variations in the mode of combustion of the volatile fractions. If the kinetic studies are correct then the fuel NO formation can be reduced by reducing the oxygen content of the volatile flame region. If the volatile gases burn in the wake of a series of particles as a premixed flame it can be assumed that the conversion of fuel nitrogen to fuel NO will be high. If, however, the volatile fractions burn in an envelope surrounding the coal jet it can be assumed that minimum conversions will occur when the oxygen for combustion approaches the flame envelope from one side only. This means that the primary fuel jet does not contain oxygen.

8.2 The Influence of Burner Variables on Nitric Oxide Formation in Pulverized Coal Flames

8.2.1 Burner Conditions Associated with Maximum Emissions

In Section 8.1 the emission of nitric oxide from pulverized fuel flames was attributed primarily to the formation of fuel NO from the volatile fuel nitrogen compounds. If this is correct then burner conditions which tend to rapidly mix the coal with all the available air will produce maximum emissions. This rapid mixing of coal and air is most efficiently achieved in high intensity Type II flames. In the parametric investigations described in Section 6, three fuel injectors produced Type II flames:

- low primary velocity axial fuel injectors at medium swirl levels;
- annular fuel injectors at medium swirl levels;
- radial fuel injectors at all swirl levels.

Figs. 8.3 and 8.4 show the emission characteristics of radial and divergent injectors for both the high nitrogen and the low nitrogen coals. The following features can be seen in these results:

- nitric oxide emissions are always higher with the high nitrogen coal;
- swirl has almost no effect on nitric oxide formation in flames produced with the radial fuel injector but causes an increase in the emissions from the annular injectors.
- emissions with both injector types are almost independent of the primary air supply.

The radial fuel injector rapidly mixes the coal and air at all swirl levels, because mixing is rapid then it is immaterial whether or not the air supplied either with the primary or with secondary supply. Also rapid mixing means that the volatile nitrogen

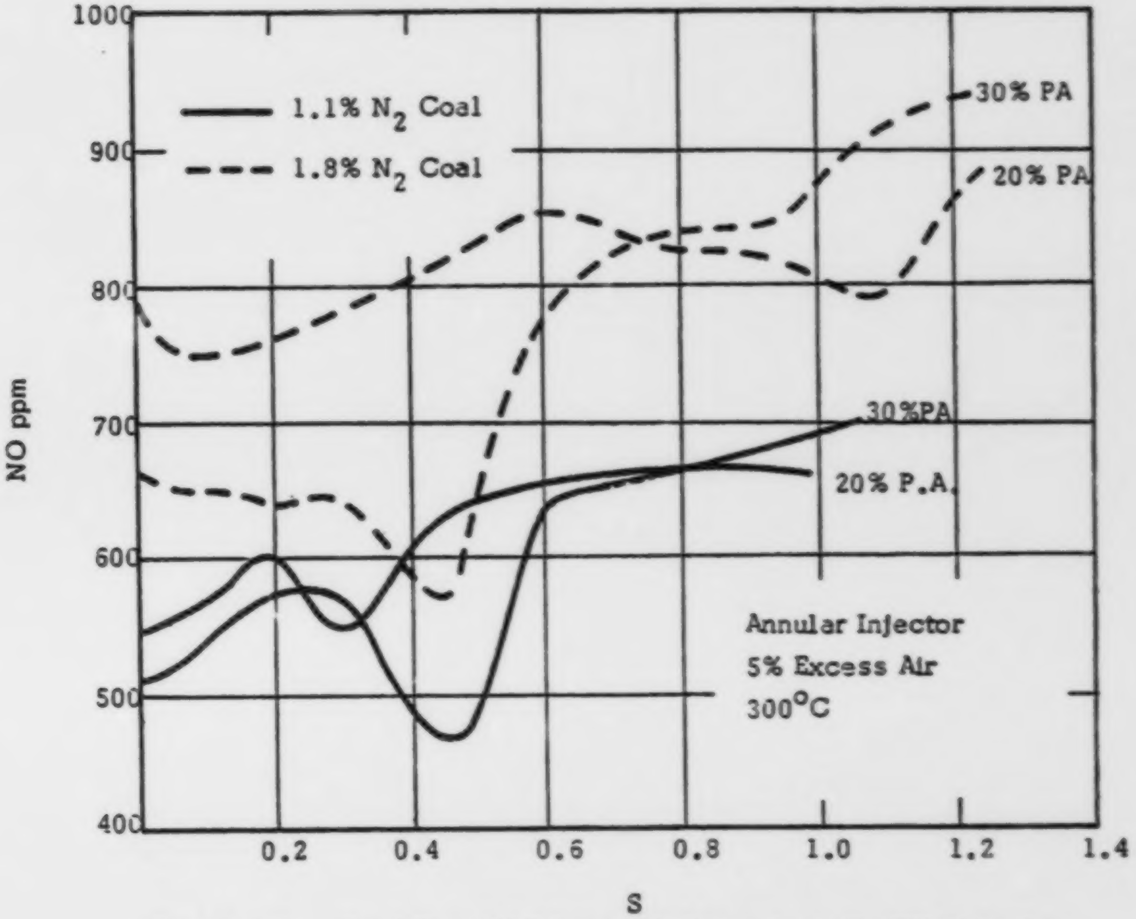

Figure 8.3 The Effect Of Primary Air Percentage On NO Emission

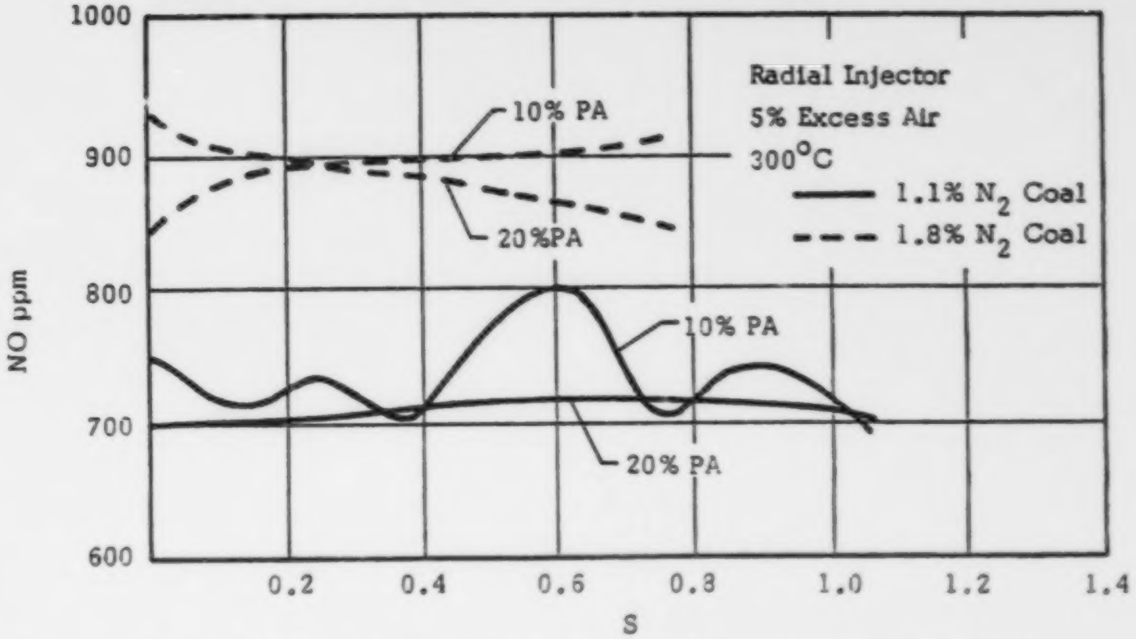

Figure 8.4 The Effect Of Primary Air Percentage On NO Emission

compounds are well mixed with all the available oxygen and conditions favor reaction (16), thus fuel NO formation is a maximum. Similar conditions are achieved with the annular fuel injector at high and medium swirl levels when a normal Type II flame is produced. Mixing between fuel and air will always take place rapidly with this type of flame. It is this characteristic which enables high combustion intensities and short burnout times to be achieved plus early stable ignition within the burner divergent. Therefore, these characteristics will be required of practical burners for wall-fired boilers and emissions will tend to be high and of the same order of magnitude as those shown in Figs. 8.3 and 8.4.

8.2.2 Reductions of Nitric Oxide Emissions Associated with the Stabilization of the Ignition Front at the Injector

During the parametric investigations with axial fuel injection it was observed that nitric oxide emissions made a step decrease when the ignition front was established at the burner when the swirl level was increased. This corresponded to the conversion of a lifted flame to a jet flame with an ignition front stable at the injector. The data presented in Fig. 8.5 confirms that nitric oxide emissions are reduced when a flame zone surrounds the coal jet. These measurements were obtained acidentally. Initially, fuel injector A was uncooled and at high swirl levels with the ignition front stable on the injector a coating of red hot coke was deposited on the thick interface. This char acted as an ignition source so that when the swirl was reduced to zero the ignition zone remained stable at the injector. Figure 8.5 shows the measured flue gas nitric oxide concentration as the swirl level is increased to a maximum and then decreased. The coal jet was always enclosed by a flame zone as the swirl was reduced to zero.

A significant difference only exists between the measured concentrations with increasing and decreasing swirl at low swirl levels. At zero swirl with the ignition front some distance from the injector the emission was 650 ppm. This was reduced to 380 ppm when the entire coal jet was surrounded by a region of combusting volatiles. It was stated earlier that high oxygen concentrations associated with the volatile combustion will preomote fuel NO formation.

In the lifted flame the ignition distance was approximately 0.75 m. Thus at the ignition point the primary and secondary jets are well mixed, the axial coal

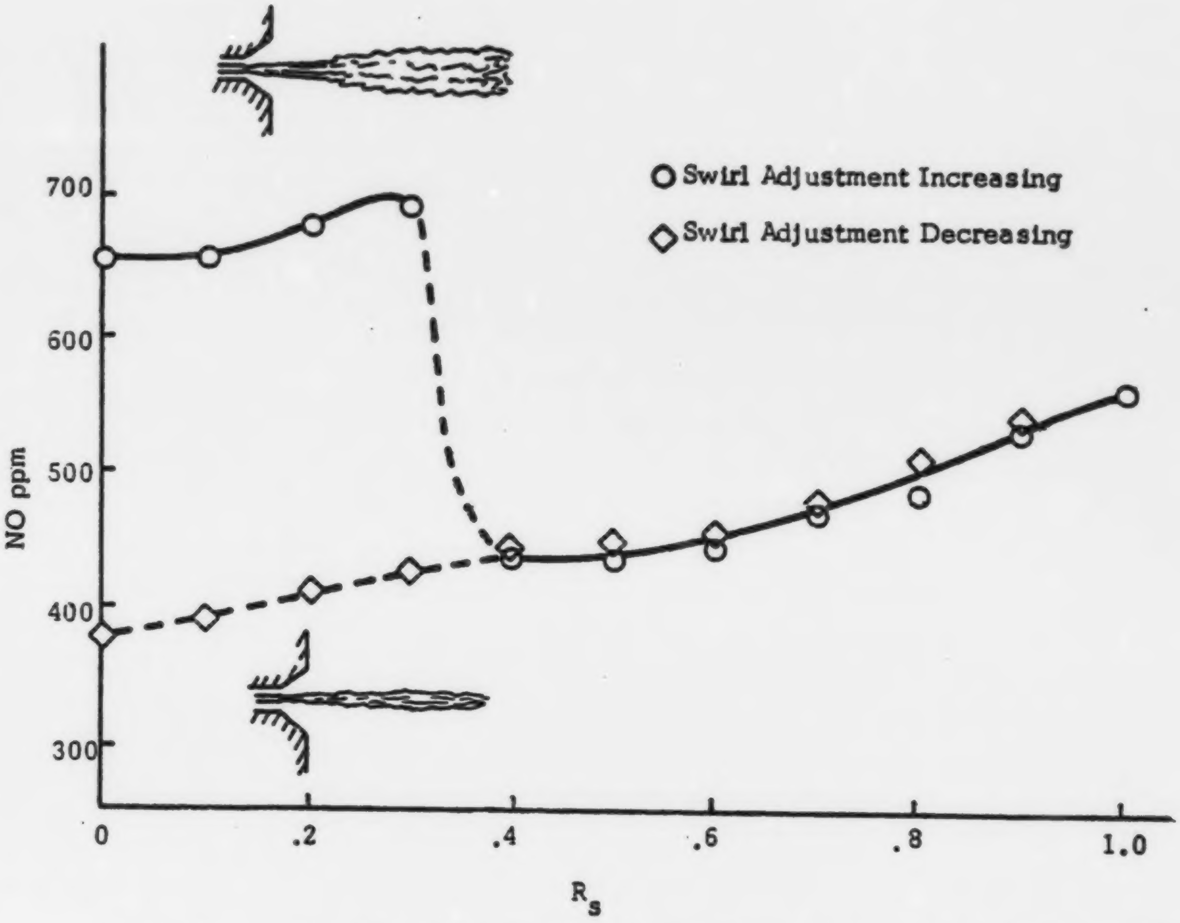

Figure 8.5 The Effect On Nitric Oxide Emission Of An Ignition Front Surrounding The Coal Jet (Injector A, Throat Of 35° Quarl, 20% Excess Air 20% Primary Air)

concentration reduced and the axial oxygen/coal ratio increased. Consequently, when the volatile gases begin to burn, not only is oxygen already mixed with volatile fractions, but oxygen can mix into the ignition front from both boundaries. When an injector stabilized ignition front is formed only that oxygen originally supplied as primary air is mixed with the fuel jet. Therefore, the oxygen/coal ratio will be reduced and less oxygen will be mixed with the volatile fractions prior to ignition. The presence of the ignition front acts as a semi-permeable barrier preventing further fuel/air mixing before the volatiles have been burned.

8.2.3 Burner Conditions for Minimum Nitric Oxide Emissions from Pulverized Coal Flames

If the preceding discussions contain any truth, then the burner conditions which will give minimum emissions can be specified:

- the coal jet should contain the minimum quantity of primary air;
- mixing between the primary and secondary streams should be avoided;
- the ignition zone for the volatile fractions should be formed as close to the injector as possible.

These conditions are best fulfilled by using a high velocity, single hole axial injector with the minimum amount of primary air. The design of the experimental system used during the furnace investigation did not allow the primary air percentage to be varied independently. Any variation in the primary air supply to a particular injector also resulted in a change in both primary and secondary velocities. To overcome this problem three fuel injectors (B, C and H) were used, which had similar primary velocities with different quantities of primary air. Thus the effect of the primary air percentage on nitric oxide formation could be investigated without interference from variations in other parameters, particularly the primary velocity. The effect of the primary air percentage on the emission curves for three different quarl-injector position arrangements can be seen in Figs. 8.6, 8.7 and 8.8.

In general, the emission of NO decreases as the swirl intensity increases, this is particularly evident for the parallel burner exit. This is attributable to dilution of the combustion air with external recirculation prior to mixing with the

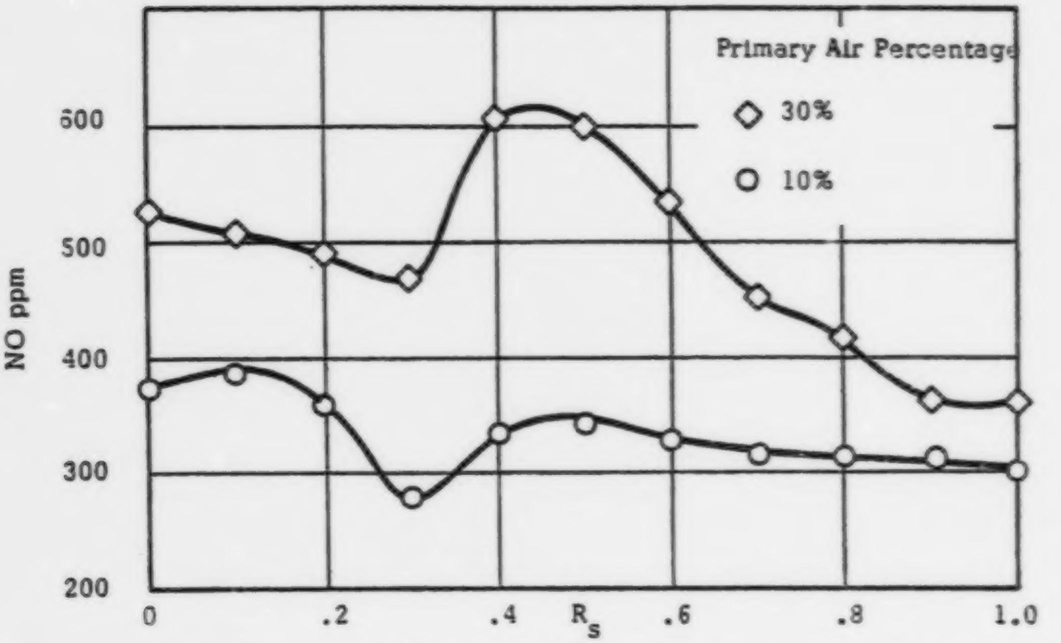

Figure 8.6 The Influence Of Primary Air Percentage With Injector In The Throat Of A 25° Divergent

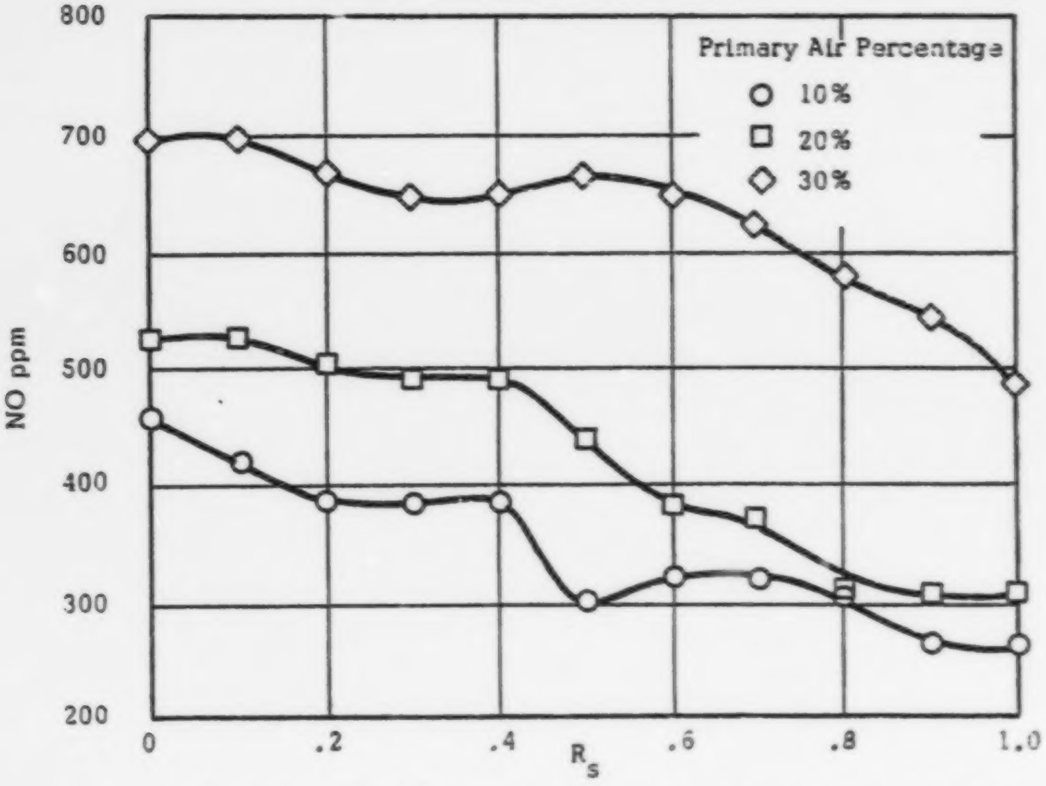

Figure 8.7 The Influence Of Primary Air Percentage With Injectors At The Exit Of A 0 Divergent

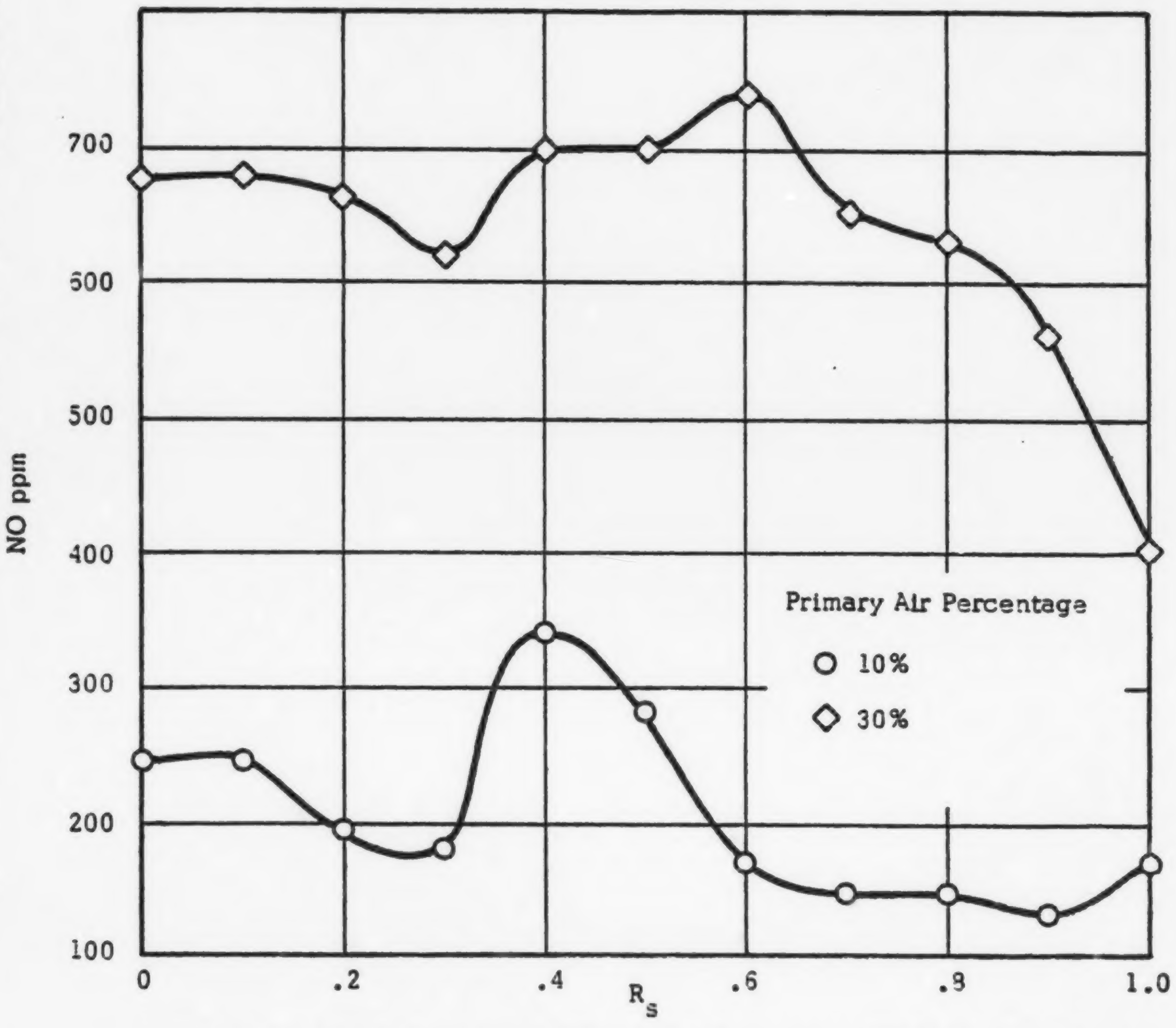

Figure 8.8 The Influence Of Primary Air Percentage With Injectors At The Exit Of A 25° Divergent

Burner Conditions Concerning Figures 8.6, 8.7, and 8.8

Primary Air Percentage	Fuel Injector	Primary Velocity
10%	H	52 m sec^{-1}
20%	C	52 m sec^{-1}
30%	B	57 m sec^{-1}

fuel. At the high primary velocities heat release occurs at some distance from the burner throat and primary/secondary mixing is delayed.

The results obtained with the high velocity, single hole injectors are in agreement with the assumptions concerning nitric oxide formation in pulverized fuel flames. Minimum emissions occur with minimum primary air percentage provided primary/secondary mixing is limited to a minimum before volatile combustion is initiated. The influence of divergent angle and fuel injector position can be seen in Figs. 8.9 and 8.10. Optimum conditions appear to occur when the fuel injector is placed at the exit of a 25^{o} quarl. This may be the optimum condition because:

- fuel injection at the exit will mean that combustion air will entrain more inert products prior to combustion. This will both lower oxygen concentrations and also reduce thermal NO formation which may now be significant at these low emission levels;
- ignition stability at the injector will require mixing at the jet boundary between hot products produced upstream. A divergent exit of 25^{o} is optimum for the strength of the internal recirculation zone.

It must be clearly stated that although these minimum emissions can be obtained, the flames which produce them are jet flames and not high intensity Type II flames, and as such, may not be suitable for industrial application. Also, high swirl levels and primary velocities require the acceptance of larger burner pressure drops than those used at present.

Two flames measured in detail during the furnace investigation also provided further evidence to support the hypothesis that an axial jet flame with minimum oxygen/coal ratio in the primary stream produced minimum NO. Figures 8.11 and 8.12 allow a comparison to be made between the axial distribution of temperature and species concentration.

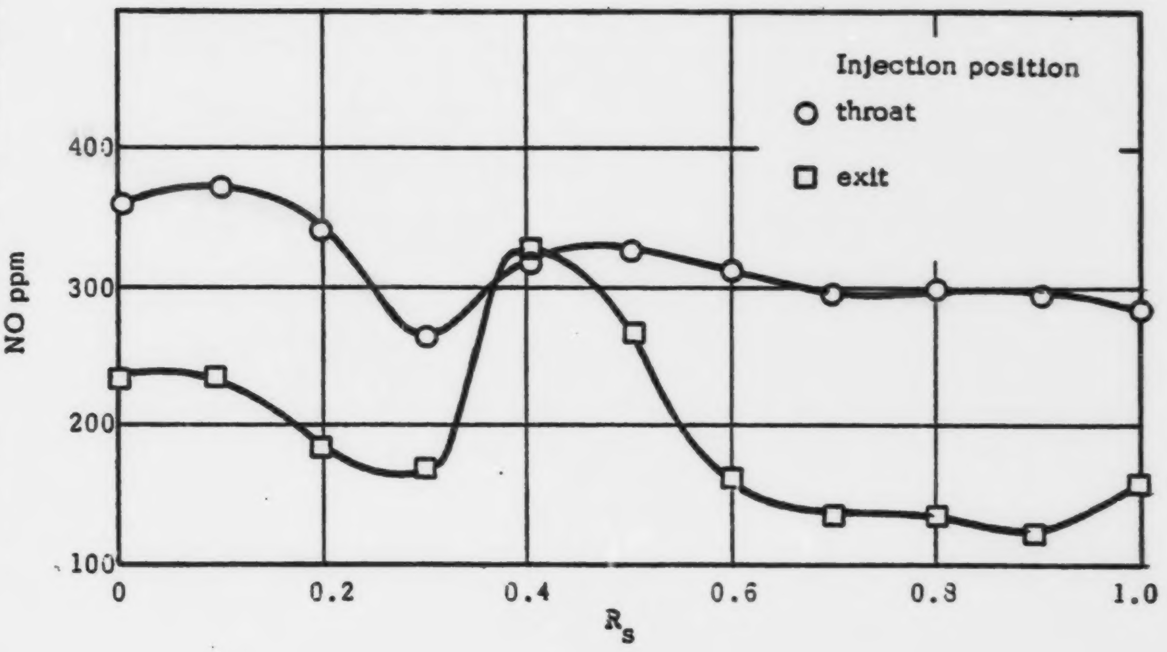

Figure 8.9 Effect Of Injection Position (Injector H, Throat 176 mm Diam., 5% Excess Air, 300°C Preheat, 10% Primary Air)

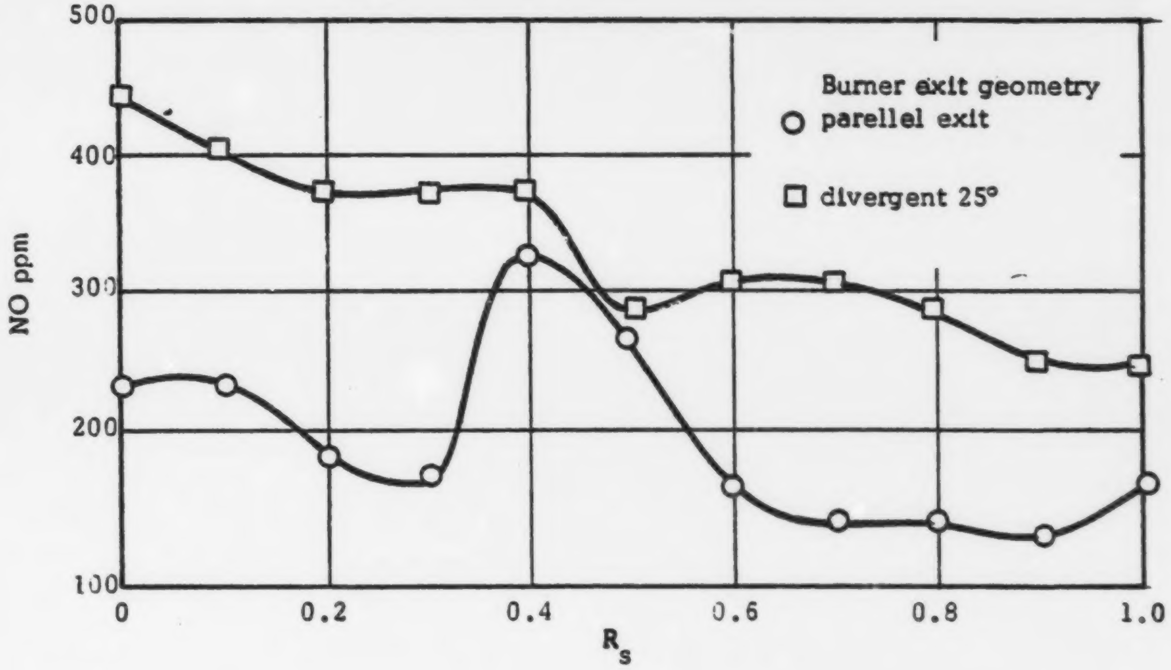

Figure 8.10 Effect Of Burner Exit Geometry (Injector H, At Exit Throat 176 mm Diam., 5% Excess Air, 300°C Preheat, 10% Primary Air

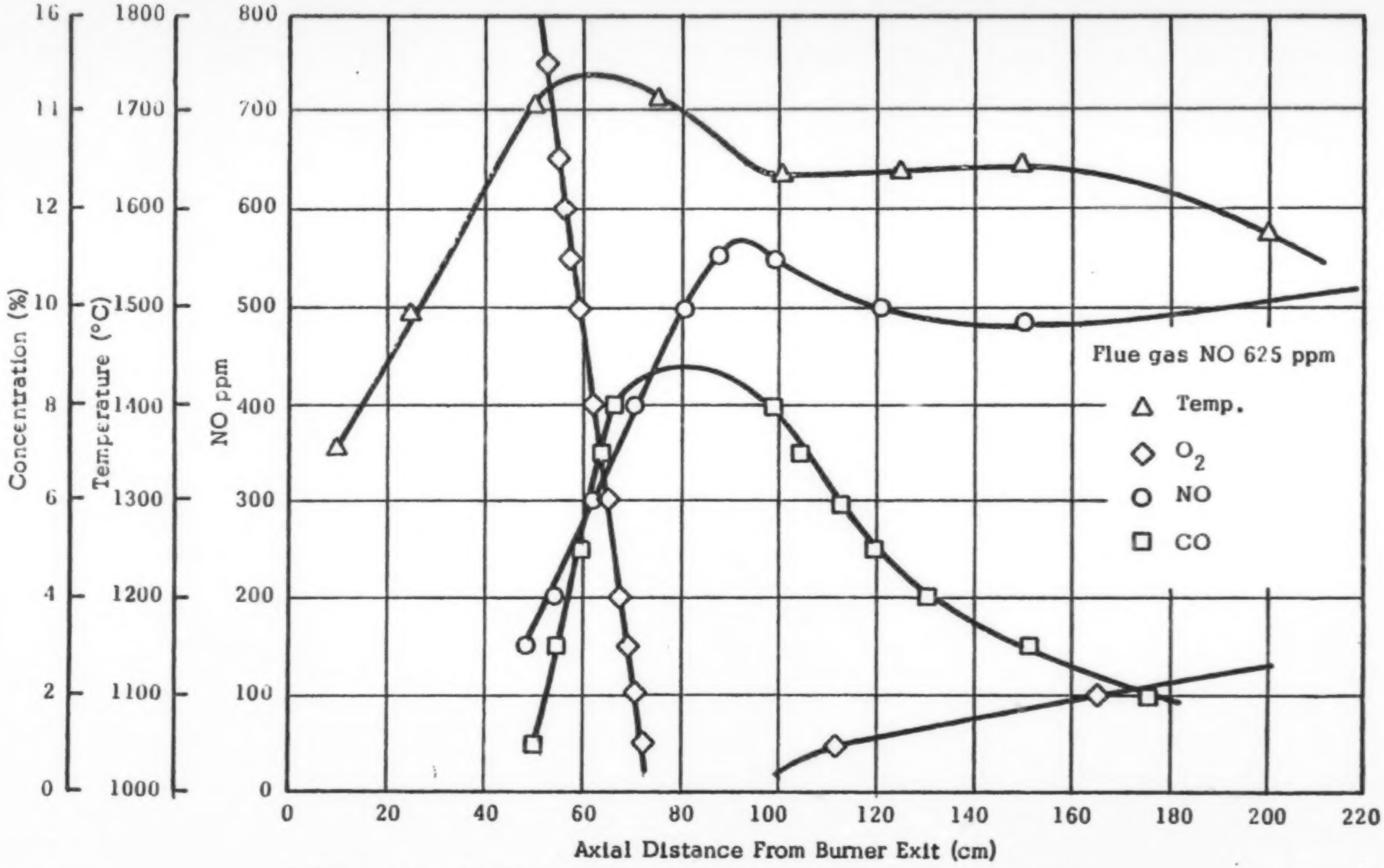

Figure 8.11 Axial Temperature And Species Distribution, Flame 123

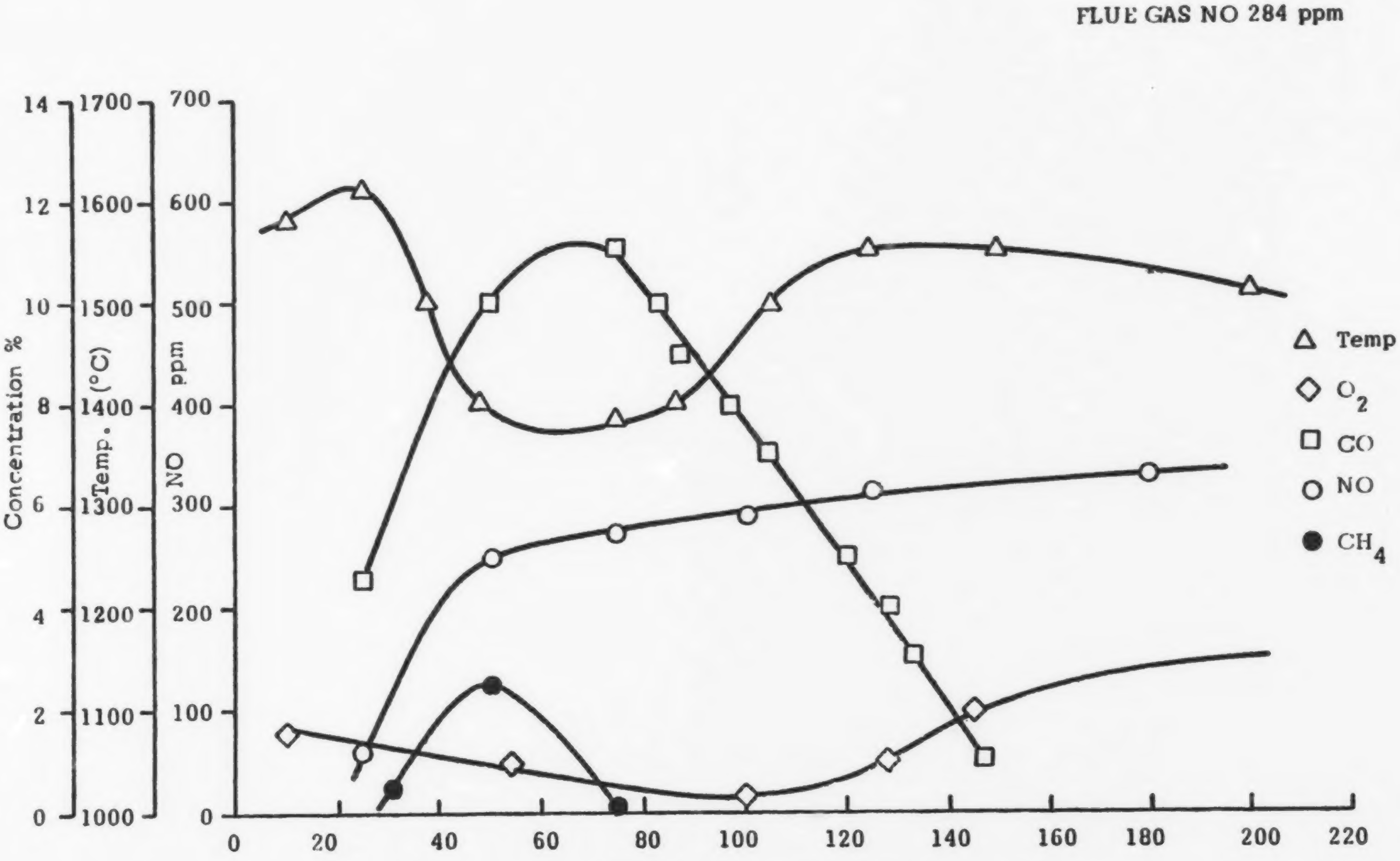

Figure 8.12 Axial Temperature And Species Distribution, Flame 124

The only difference between the two flames was the amount of primary air content. It can be seen that in the flame with the lower final emission and primary air percentage the axial oxygen concentration drops rapidly to zero. High concentrations of carbon monoxide and hydrogen also occur with the lower primary air flame and significant quantities of methane were also detected on the axis. These measurements confirm that low oxygen/coal ratios within the primary jet reduce NO formation.

8.2.4 The Influence of Primary Velocity on Nitric Oxide Formation

One of the burner variables which were specified as being necessary for minimum nitric oxide emission characteristics was a high primary velocity. The influence of the primary velocity on the emission characteristics of pulverized fuel flames can be judged from the results presented in 8.13 and 8.14. Fuel injectors B, C, and H had primary velocities of 19, 26, and 52 msec^{-1} respectively, and the secondary velocity was 41 msec^{-1}.

At zero swirl with the injectors both at the throat and at the exit the emissions were in the order:

- B greater than H greater than C; however, at higher swirl values the emission increases with decreasing primary velocity. The anomolous behavior at zero swirl is probably due to a combination of different primary/secondary mixing rates and ignition distance.

The effects of primary velocity are more pronounced when the fuel is injected from the throat of the burner divergent (Fig. 8.13). The form of the emission curves is associated with the transition from one high intensity flame type to another. In Section 3 the conditions necessary to form Type I and Type II flames were discussed, and it was stated that the parameter G, ratio of the primary to secondary velocities has a strong influence on the type of flame produced. The results presented in Fig. 8.13 are for a fixed secondary velocity and variable primary velocity. The emission characteristics are attributable to:

High Primary Velocity (Injector H)

The high primary velocity ensures that a reverse flow zone cannot form on the axis and an injector stabilized flame is produced. The interaction between the annular reverse flow zone and the primary jet gives ignition stability but does not entrain sufficient fuel to form the bulbous base characterisics of Type I flames and the emission curve is almost flat.

Medium Primary Velocities (Injector C)

Even at low swirl intensities, the primary jet velocity is insufficient to penetrate the axial reverse flow zone. The fuel jet is split, a Type II flame is produced and the emission increases. The reason for the high emissions with Type II flames has been explained earlier. The high combustion intensity within the divergent due to increasing swirl alters the static pressure distribution and the primary velocity is sufficient to allow the fuel jet to penetrate some way into the reverse flow zone. However, complete penetration does not occur and a Type III flame is produced. Mixing between primary jet and secondary air is restricted and the amount of nitric oxide produced is reduced.

Low Primary Velocities (Injector B)

Type II flames are more easily produced and better fuel/air mixing is possible at lower swirl levels. Thus emissions are higher than with injector C. Only at high swirl numbers is it possible to produce partial Type III flames which quickly form Type II flames as the swirl is increased.

The difference between the emission curves presented in Figs. 8.13 and 8.14 is due to the modifying influence of the position of the injector on swirl. With the injector at the exit of the burner, the primary jet does not experience the full effects of the axial pressure gradients and thus, the primary/secondary mixing is less intense. Consequently, even with the low velocity injector B, a Type II flame is not produced.

8.2.5 The Influence of Throat Velocity on Nitric Oxide Formation

In natural gas flames an increase in throat velocity invariably produced a decreased emission. In Section 7 this effect was attributed to a reduction in thermal NO formation. If thermal NO formation is discounted in coal flames, then the primary effect of throat velocity will be to change the rate of primary/secondary mixing or to reduce the oxygen concentration of the primary jet by entrainment of combustion products. The results are confused and the effect of throat velocity is obviously dependent on other burner parameters, e.g.,

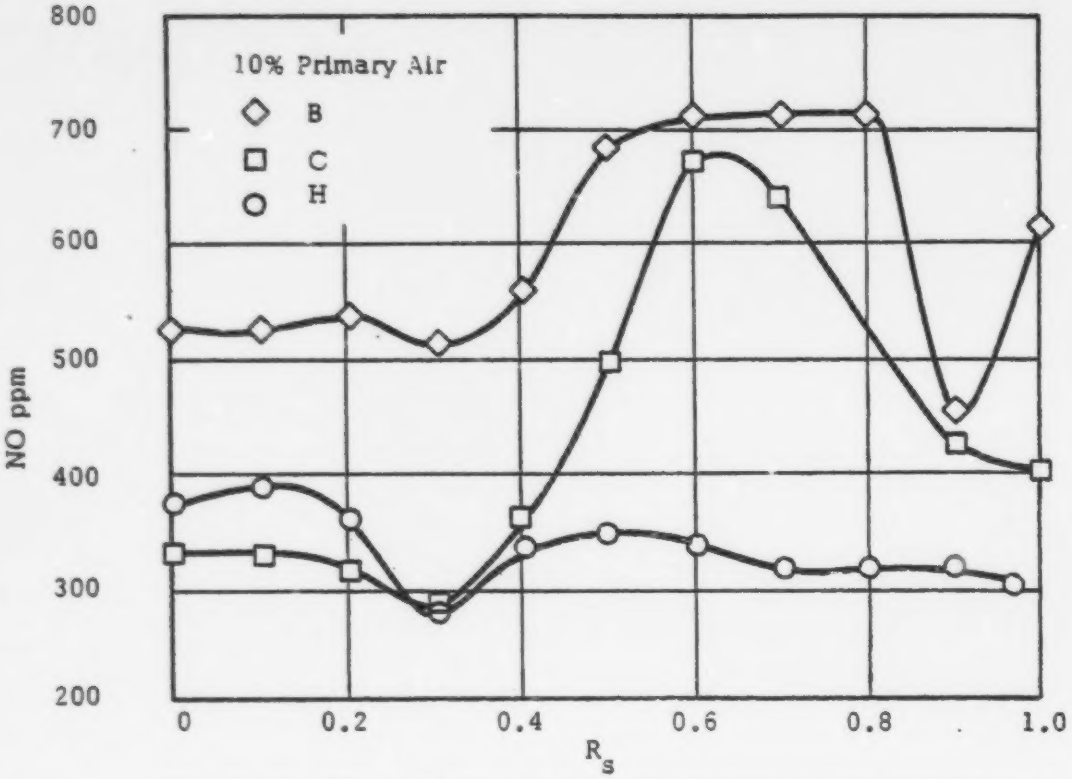

Fig. 8.13 The Influence of Primary Velocity on NO Formation in PF Flames (Injector in the Throats)

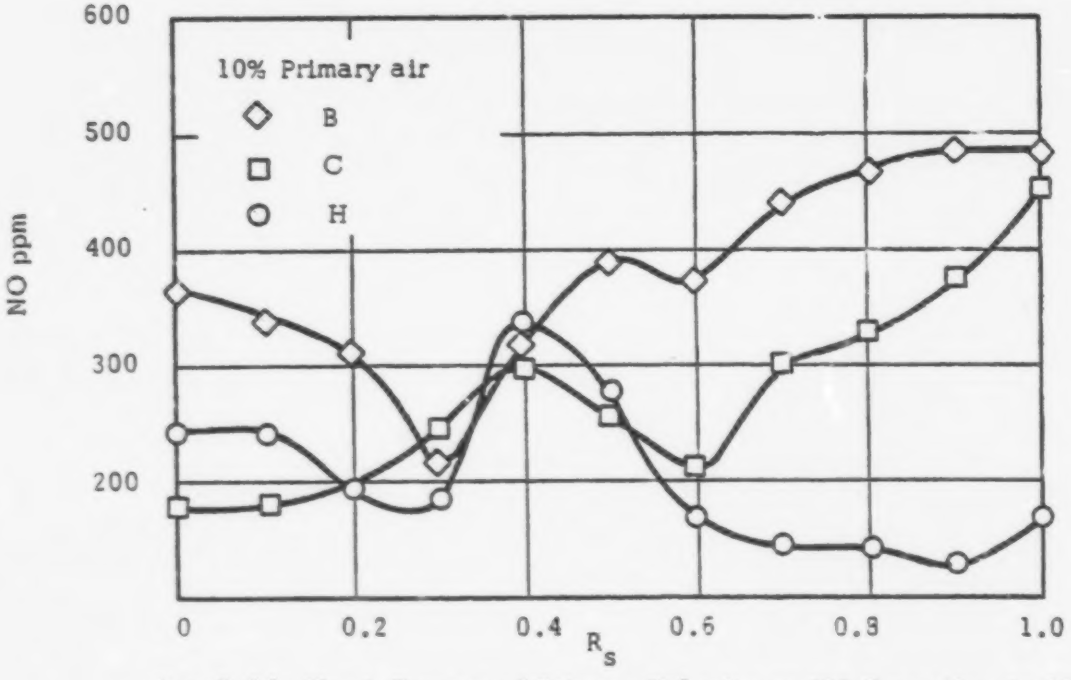

Fig. 8.14 The Influence of Primary Velocity on NO Formation in PF Flames (Injector at the Exit of the Divergent)

injector type, divergent angle and primary air percentage. The results obtained during the parametric investigations covered the entire range; increased throat velocity either increases, decreases or has no effect on the emission of nitric oxide.

8.3 The The Influence of Coal Type

Two different coals were used during the parametric investigations to determine whether or not the influence of burner parameters on nitric oxide formation in pulverized coal flames are dependent on coal type. The results presented in Figs. 8.3 and 8.4 confirm that the increase of emission is not directly proportional to the increased nitrogen content. However, the form of the emission curves is similar for both coals. This type of behavior was typical of all cases. The results presented in Figs. 8.15 and 8.16 show cases in which the higher nitrogen coal gave lower values and in which the difference in the emission levels was greater than that shown in Figs. 8.3 and 8.4

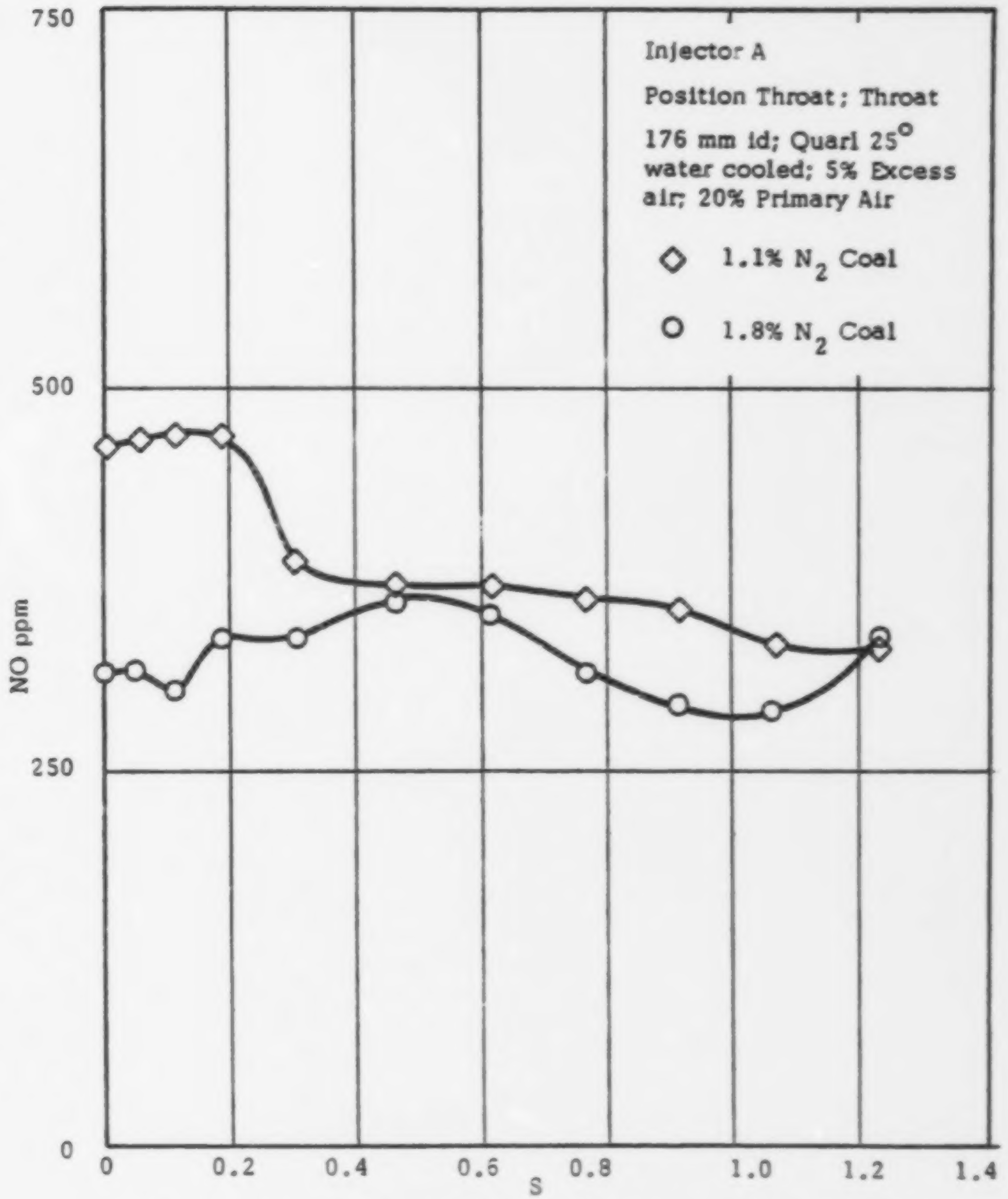

Fig. 8.15 The Influence of Coal Nitrogen Content on NO Formation

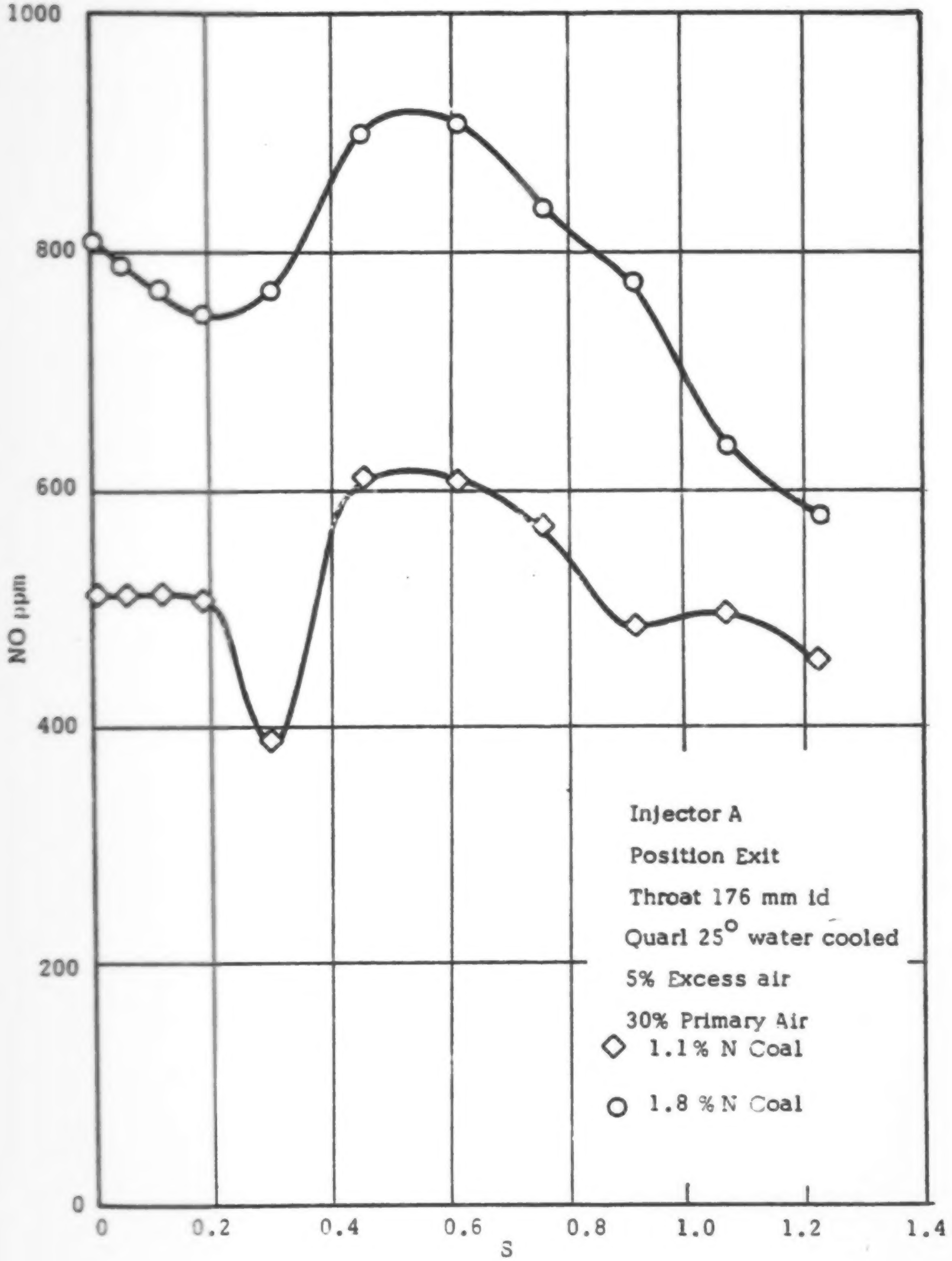

Fig. 8.16 The Influence of Coal Nitrogen Content on NO Formation

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9. CONCLUSIONS

The test program demonstrated that burner variables can significantly affect NO formation in natural gas and pulverized fuel flames and provided insight into the causative phenomena.

9.1 Natural Gas Flames

The degree of swirl was the most significant burner variable exercised in the natural gas test program. In simple jet flames, swirl was found to reduce emission of NO by increasing entrainment, thus, resulting in dilution of the reactants and a more rapid cooling of the products. For swirl levels large enough to produce internal recirculation (Type I flames), emissions were found to increase with increasing swirl, apparently because fuel was entrained and reacted at the base, thus reducing reactant dilution. In Type II flames the effect of swirl was dependent on burner geometry. With a divergent injector, increased swirl resulted in decreased emissions because of increased cooling rate for the product. The effect of swirl was less pronounced with a radial injector.

Other burner variables which had significant effects on NO production were air velocity, burner divergent angle, and fuel injector position. Increased air velocity tended to decrease emissions in all cases. The other two factors had varying effects depending on swirl level and exact geometry.

9.2 Pulverized Fuel Flames

The test results supported the theory that the most significant source of emissions in p.f. flames is the fuel NO produced in combustion of the volatile fractions of the coal. A review of the test data produced the following generalizations:

1. Low coal concentrations in the primary stream during the initial stages of heat release promote nitric oxide formation.
2. High mixing rates between fuel and air promote nitric oxide formation.
3. Any tendency to spread the coal jet promotes nitric oxide formation.
4. High primary velocities tend to reduce nitric oxide formation.
5. The influence of throat velocity is not well-defined.
6. For every burner condition there is an optimum primary air percentage. In many instances this optimum appears to be around 20 percent of the stoichiometric requirement.

7. Ignition stability at the injector can reduce emissions with axial fuel injection.
8. Maximum emissions are higher with higher fuel nitrogen contents. However, minimum emissions appear to be independent of coal type.

Caution must be applied with all these generalizations since there are many exceptions.

It can seen that many of the conditions that tend to provide the high intensity combustion and rapid burnout desired for practical systems also produce high NO emissions. Subsequent work on p.f. burner designs (described in Volumes II and III of this report) was oriented toward maintaining the desired flame characteristics while reducing the initial rate of fuel/air mixing.